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ABSTRACT

This report summarizes the results of unclassified research and development contracts in the field of peaceful use safeguards regarding the use of nuclear material. These summaries indicate there is really no sharp line of demarcation between research for safeguards and research for many other purposes. It includes areas of research effort and expenditures since the formal beginning of research for safeguards in 1955. The reports have been grouped according to the following subjects: analytical methods; calculation of heavy isotope content of reactor fuel; fuel element analysis; gamma spectrometers; inspection procedures; inventory control; mathematical models for fuel cycle analysis; plant protective and monitoring systems and devices; process instrumentation; reactivity measurements; and sampling. (Author/JG)

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RESEARCH and DEVELOPMENT for SAFEGUARDS

DECEMBER 1, 1968

Office of Safeguards and Materials Management

UNITED STATES ATOMIC ENERGY COMMISSION

SE 010 614

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Safeguards and Nuclear
Materials Management
(SPECIAL)

RESEARCH AND DEVELOPMENT FOR SAFEGUARDS

Guy M. Inman

December 1, 1968

Office of Safeguards and Materials Management

UNITED STATES ATOMIC ENERGY COMMISSION

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ABSTRACT

This document indicates the present status of active research and development programs for safeguards and summarizes the results of completed contracts. Available information concerning unclassified contracts awarded from the beginning of safeguards activities in 1955 until October 1, 1968 is included. The active contracts are being supported by the ACDA, BMwF, IAEA and the USAEC.

In addition to serving as a historical review of safeguards research and development, the document also includes the "period of contract" and contract price so that the direction of effort and the increasing expenditures can be clearly seen.

Research organizations should be able from the summaries presented to identify areas in which they have special technological competence to contribute to the new and expanding field of peaceful use safeguards.

TABLE OF CONTENTS

Page

I. INTRODUCTION 1

II. SPONSORING ORGANIZATIONS 3

III. SUBJECT INDEX 5

IV. SUMMARIES 7

V. SPONSORING ORGANIZATION INDEX
ACDA, BMwF, IAEA, USAEC 265

VII. EXPENDITURES FOR SAFEGUARDS RESEARCH 276

VIII. BASIS AND BACKGROUND OF SAFEGUARDS 279

I. INTRODUCTION

This report summarizes the results of unclassified research and development contracts in the field of peaceful use safeguards. (A brief history of the basis and background of safeguards is given in Section VIII, page 279.) All available information regarding contracts awarded from the beginning of safeguards activities in 1955 until October 1, 1968 is included.

The Atomic Energy Act of 1946 required that all special nuclear material be owned by the United States Government. At that time a system of control over the material was established to assure, to a high degree, the detection of any loss or diversion. By material assay and accounting procedures, the system demonstrated how much material should be at any location and how much was actually on hand. As indicated in Section VIII, the Act of 1946 was extensively modified in 1954 to permit the distribution of special nuclear material to private persons. The 1954 Act also provided, in the context of agreements for cooperation between the United States and other governments, for safeguards on special nuclear material distributed abroad.

In 1957, the Statute of the International Atomic Energy Agency became effective. It specifically required that safeguards be imposed on nuclear materials used in certain international activities.

The first control efforts of the USAEC after 1954 were concerned with activities collectively known as 'nuclear materials control' which were primarily for material accountability and financial purposes. To support these activities, the USAEC awarded contracts for the development of accounting procedures and analytical methods to assay the nuclear materials.

With the establishment in the USAEC of the Office of Safeguards and Nuclear Materials Management in 1967, safeguards have emerged as a more precisely defined area of endeavor aimed at preventing and detecting the unlawful diversion of nuclear materials. While this encompasses all the activities required for nuclear materials management, it includes new equipment and techniques, discussed in this document, such as tamper-proof seals and instrumentation, methods of material tagging, identification and assay, and systems studies of the entire nuclear fuel cycle.

A few research projects are reported which, although not performed for the specific purpose of safeguards, are related to safeguards.

These summaries indicate there is really no sharp line of demarcation between research for safeguards and research for many other purposes. The project summaries indicate very clearly that safeguards encompasses such diverse disciplines as radiation chemistry, accounting, statistics, reactor physics, computer programming, and operations analysis as well as efforts in the legal, financial and political areas.

This is a specialized report prepared for the information of those concerned with safeguards and nuclear materials management. It is not intended to include the technical detail that is presented in topical, or even progress reports. It supplements "Nuclear Science Abstracts" and other abstracting documents in that it also reports work in progress. The "Period of Contract" and the contract price are included to indicate the direction and scope of safeguards R&D; such information is not usually available from technical reports and published abstracts.

The original objective of this document was to summarize the results of individual contracts which had produced topical reports covering specific areas of investigation. In most cases this approach has been satisfactory. However, certain large contracts cover such a variety of subjects that several topical reports were prepared for a single contract. In this case, summaries of the individual reports rather than the contract are given (see page 122).

One purpose of this report is to indicate the areas of research effort and the expenditures since the formal beginning of research for safeguards in 1955 with the awarding of a contract by the USAEC for an inventory control study (see page 145).

In order to most effectively direct the expenditure of funds for safeguards R&D, it is necessary that the Office of Safeguards and Materials Management be cognizant of all work in this area. One purpose of this summary document is to indicate, as mentioned above, the historical development of safeguards R&D and to establish a perspective so that areas requiring effort can be identified.

The report summaries have been grouped according to subject as much as possible. The subject matter of some reports would permit their being put in any of several subject categories; these reports have been arbitrarily assigned to a suitable category.

The active research contracts are being supported by the United States Arms Control and Disarmament Agency, the Federal Ministry for Scientific Research (Federal Republic of Germany), the International Atomic Energy Agency and the United States Atomic Energy Commission.

II. SPONSORING ORGANIZATIONS

A. ACDA - United States Arms Control and Disarmament Agency
Department of State
Washington, D. C. 20451

B. BMwF - Bundesministerium für wissenschaftliche Forschung
(Federal Ministry for Scientific Research)
Federal Republic of Germany
532 Bad Godesberg, Germany

C. IAEA - International Atomic Energy Agency
Vienna, Austria

D. USAEC- United States Atomic Energy Commission
Office of Safeguards and Materials Management
United States Atomic Energy Commission
Washington, D. C. 20545

III. SUBJECT INDEX

Page

A. Analytical Methods 7

B. Calculation of Heavy Isotope Content of Reactor Fuel 35

C. Fuel Element Analysis 51

D. Gamma Spectrometers 111

E. Inspection Procedures 117

F. Inventory Control 143

G. Mathematical Models for Fuel Cycle Analysis . . 163

H. Miscellaneous 183

I. Plant Protective and Monitoring Systems and Devices . 205

J. Process Instrumentation 223

K. Reactivity Measurements 247

L. Sampling 259

IV. SUMMARIES

Page

A. Analytical Methods

1. Selected Measurement Methods (USAEC) 9
2. Interlaboratory Evaluation Program (USAEC) 10
3. Standard Reference Materials (USAEC) 11
4. Analytical Procedures and Facilities for a Nuclear Materials Control System (USAEC) 12
5. Gamma Scan Measurement of Air Filters (USAEC) 13
6. Development of Primary Standards for Plutonium + Uranium (USAEC) 14
7. Calorimetric Assay of Radioactive Materials (USAEC) 15
8. Plutonium Content by Calorimetry (BMwF) 18
9. Plutonium Nitrate Shipping Solution Measurements (USAEC) 19
10. Plutonium Oxide Shipping Measurements (USAEC) 21
11. Plutonium Compound Evaluation Program (USAEC) 24
12. Analyst Performance on Synthetic Purex Solutions (USAEC) 25
13. Qualification of Umpire Laboratories (USAEC) 26
14. Isotopic Composition Analysis by X-ray Fluorescence (BMwF) 31
15. Isotopic Composition Analysis by Isotopic Dilution Spectrometry (BMwF) 32
16. Isotopic Composition Analysis by Photometry (BMwF) 33
17. Rapid Process Control of Alpha Emitting Isotopes (USAEC) 34

1. Research Contract No. $

Title

Selected Measurement Methods

Institution: Several AEC Laboratories and AEC Contractors

Principal Scientific Investigators: R. J. Jones, USAEC

Period of Contract: 1959-1963

Results

The result of this program was the publication of a 419 page analytical manual which includes selected analytical methods for plutonium and uranium along with discussions of sampling, bulk measurement techniques and the use of standard reference materials.*

Costs were not separated for this program. The program was started in 1959 and was conducted during the normal course of laboratory operations. It incorporates the efforts of many persons in the field who contributed to the publication.

The manual is organized as follows:

Part I GENERAL CONSIDERATIONS

Chapter 1	Introduction
Chapter 2	Standard Reference Materials
Chapter 3	Sampling
Chapter 4	Bulk Measurements
Chapter 5	Statistics of Measurement

Part II ANALYTICAL METHODS

Uranium Concentration Measurement
Uranium Isotopic Composition Measurement
Plutonium Concentration Measurement
Plutonium Isotopic Composition Measurement
Supplementary Methods

*"Selected Measurement Methods for Plutonium and Uranium in the Nuclear Fuel Cycle" 1963 Jones, Ralph J. (USAEC) (TID-7029) (For sale by the Superintendent of Documents, U. S. Government Printing Office, Washington, D. C. 20402)

2. Research Contract No. $ 36,000

Title

Interlaboratory Evaluation Program

Institution: USAEC New Brunswick Laboratory

Principal Scientific Investigators: C. J. Rodden, M. W. Lerner

Period of Contract: 1965

Results

This program, begun by the Division of Nuclear Materials Management in 1965, evaluated the methods in TID 7029, "Selected Measurement Methods for Plutonium and Uranium in the Nuclear Fuel Cycle." The evaluation was accomplished by analysis of a series of samples prepared and distributed by NBL to thirty-four different laboratories throughout the U. S.

3, Research Contract No. $ 1,950,000

Title

Standard Reference Materials

Institution: USAEC New Brunswick Laboratory (NBL)
National Bureau of Standards (NBS)

Principal Scientific Investigators: C. J. Rodden (NBL); J. K. Taylor and J. L. Hague (NBS)

Period of Contract: 1957 to 1963 (see note)

Results

This program, initiated in 1957 by the Division of Nuclear Materials Management, has resulted in the development, preparation and certification by NBS of the following primary standards:

a. A series of 16 isotopic standards for uranium ranging from 0.5% U-235 to 93% U-235.

b. An isotopic plutonium standard having about 90% Pu-239 and about 8% Pu-240.

c. Chemical standards for both uranium and plutonium.

The program since 1963 has been funded by NBS but has continued to be an AEC-NBS program. Presently under development are:

a. Three additional primary uranium isotopic standards, natural, highly depleted and highly enriched.

b. Additional plutonium isotopic standards of higher Pu-240 content.

c. Working standards for chemical analyses for both plutonium and uranium.

Note: This is a continuing program which has been funded by the National Bureau of Standards since 1964.

4. Analytical Procedures and Facilities for a Nuclear Materials Control System (12/1/59) WCAP-6032

C. C. Thomas, Jr.

Analytical procedures and facilities suitable for use in a Nuclear Materials Control System (NMCS) are specified. Techniques, precisions, and accuracies are considered for the analysis of various process samples for uranium, plutonium, free acid, specific gravity, and other data. The handling and storage of analytical samples is also discussed, and specific recommendations are given.

5. Research Contract No.	$ 8,000

Title

Gamma Scan Measurement of Air Filters

Institution: New York Operations Office, USAEC

Principal Scientific Investigator: Ira M. Cohen, USAEC, NYO.

Period of Contract: 1962

Results

This program was designed to provide improved measurement of the U-235 content of air filters that had been used in plant air systems. The technique has proven quite useful and is being developed to more widespread use.

Started in 1962, the majority of this effort was done in the normal course of the field office nuclear materials management effort. On several occasions additional effort was required which required funding. This was provided by the Division of Nuclear Materials Management and totaled about $8,000.

6. Research Contract No. AT-33-1-GEN-53 $ 50,000
(Related to Safeguards)

Title

Development of Primary Standards for Plutonium + Uranium

Institution: Mound Laboratory, Miamisburg, Ohio

Principal Scientific Investigators:

Period of Contract: 7/1/64 to 6/30/68

Published Reports

1) Rogers, D. R. and Brown, W. B., Potassium Plutonium Sulfate, MLM-1212, Mound Laboratory, Miamisburg, Ohio (December 11, 1964), 13 pp.

2) Plymale, D. L. and Smith, W. H., "Uranyl Tropolonate Complexes", J. Inorg. Nucl. Chem., (to be published).

3) Plymale, D. L. and Smith, W. H., "Plutonium Tropolonate Complexes", J. Inorg. Nucl. Chem., (to be published).

4) Smith, W. H., "Thermal Dehydration of Uranyl Nitrate Hydrates", J. Inorg. Nucl. Chem., (to be published).

5) Plymale, D. L., "Plutonium (IV) Nitrate Complexes of Triphenylarsine Oxide and Triphenylphosphine Oxide", J. Inorg. Nucl. Chem., (to be published).

6) Silver, G. L., Separation of Uranium and Thorium from Plutonium, (patent applied for).

16

7. Research Contract No. AT-33-1-GEN-53 $ 120,000
(Related to Safeguards) FY-68

Title

Calorimetric Assay of Radioactive Materials

Institution: Mound Laboratory, Miamisburg, Ohio

Principal Scientific Investigators: B. C. Blanke, J. F. Eichelberger
K. C. Jordan

Period of Contract: 1945 to continuing

Background & Principle

Mound Laboratory has had a continuous calorimetry program since 1945. During this time, the laboratory has participated with other AEC sites on a wide variety of research, development, and production problems involving both radioactive and stable materials. In these problems, the power range from microwatts to hundred watts has been measured, and most radioactivity shipped from Mound Laboratory in millicurie or higher amounts has been calorimetered to satisfy the requirements of nuclear management.

Since the inception of the calorimeter program, over 85 calorimeters have been built for use at Mound Laboratory and other AEC sites. In early development of calorimetry the use of twin-bridge calorimeters was stressed. But, as a result of the development here of a nuclear thermoelectric generator under a U. S. Signal Corps contract, several micro-calorimeters similar in design to the thermoelectric generator using thermopiles and having detectabilities of less than one erg per second have been constructed. Recently, the need for calorimetering large samples of irregular geometry with outputs greater than several watts led to the development of a twin-bath calorimeter.

Most of Mound Laboratory's calorimeters operate near and slightly above room temperature. However, several calorimeters have been pushed to the boiling point of the water bath, and two high-temperature calorimeters have been constructed. The first high-temperature calorimeter can operate up to 650°C and the second up to 800°C. Both of these high-temperature calorimeters are of the twin-bridge type.

Results

The capabilities of calorimetric analysis techniques are indicated by the half-life measurement of Actinium-227 and Thorium-227 using twin heat flow calorimeters.

The half-life of Ac-227 and probable error was determined to be 21.7728 + 0.0029/-0.0032 years (based on an ordinary tropical year of 365.2422 days) and the half-life and probable error of

Th-227 was 18.7176 ± 0.0052 days. The Ac-227 half-life was determined on three samples in three calorimeters over a 14-year period.

With the data that have been accumulated on Ac-227, it is suggested that it be considered for use as a laboratory standard for calorimetry. It has the following excellent characteristics for this purpose:

(1) Actinium-227 is a naturally occurring isotope in a branch of the uranium-235 chain. It can be separated from irradiated Ra-226 and is becoming readily available;

(2) The half-life of Ac-227 has been accurately measured to be 21.7728 years;

(3) The half-lives of all the daughters have been accurately measured;

(4) The half-lives of the decay products are so short that, after one year decay, equilibrium is established. Further, the Ac-227 half-life is controlling after the equilibrium is established;

(5) Chemical separation of the actinium from daughter products to give an accurate t = 0 is an established process;

(6) Chemical separation of the actinium gives an initial, almost pure, beta-emitting product (approximately 2% alpha) that can be critically evaluated for the absence of all other radioisotopes; and

(7) The power output of the sample increases in 185 days to 450 times the initial output at the time of chemical separation.

The disadvantage of the shorter half-life of the actinium compared with radium is more than offset by the greater accuracy with which it is known.

Published Reports

"Calorimetric Determination of the Half-Life of Polonium-210", The Physical Review, 96, 719-721, (1954), J. F. Eichelberger, K. C. Jordan, S. R. Orr, and J. R. Parks.

"High Temperature Differential Resistance Bridge Calorimeters", Review of Scientific Instruments, 35, 875-880, (1964), T. K. Engel, K. C. Jordan, G. W. Otto, and D. M. Scott.

"Half-Life of Radium-223", J. Inorganic Nucl. Chem., 27, 1881-1887 (1965), H. W. Kirby, K. C. Jordan, J. Z. Braun, M. L. Curtis, and M. L. Salutsky.

"Half-Lives of Actinium-227 and Thorium-227 Measured Calorimetrically", *Standardization of Radionuclides*, Proceeding of a Symposium, Vienna, October 10-14, 1966, K. C. Jordan and B. C. Blanke.

"Half-Life of Tritium", *J. Inorg. Nucl. Chem.*, 29, 2129-2131 (1967), K. C. Jordan, B. C. Blanke and W. A. Dudley

8. Research Contract No. $

Title

Plutonium Content by Calorimetry

Institution: ALKEM

Principal Scientific Investigator: W. Stoll

Period of Contract: August 1967 to continuing

Background & Principle

The firm ALKEM has developed a calorimeter for determining the plutonium content in plate type fuel elements for the Reactor SNEAK within an accuracy of ± 0.2%

Objective

Extend the calorimetric analysis method to cylindrical fuel elements.

9. Research Contract No. AT(29-1)-1106 $ 157,000

Title

Plutonium Nitrate Shipping Solution Measurements

Institution: Dow Chemical Company, Rocky Flats Plant

Principal Scientific Investigators: J. T. Byrne, R. L. Delnay

Period of Contract: 1962 to 1964

Background & Principle

Plutonium nitrate solution is one of the forms in which the Atomic Energy Commission will purchase privately produced plutonium. Within the AEC complex, however, there has been a long history of differences between the shipper-determined and receiver-determined plutonium content of plutonium nitrate solutions. In many cases, these differences have been of the order of several percent. The causes for and the conditions relating to these differences had never been adequately defined. It was necessary, then, that before plutonium nitrate is used as a form for the purchase of privately produced plutonium, a thorough study be undertaken to investigate the problems associated with the shipping of plutonium nitrate. The Commission requested such a study in August of 1962.

Objectives

Determine the causes for, and eliminate if possible, the shipper-receiver differences on shipments of plutonium nitrate.

Conclusions

Shipper-Receiver Differences

Using by-difference weight bulk measurements and a standardized analytical method, an interplant shipment of six 3-liter containers of plutonium nitrate was made with an average shipper-receiver difference less than 0.2%.

In twenty intraplant simulated shipper-receiver measurements and in thirteen interplant shipments of plutonium nitrate, the relative standard deviation of the shipper-receiver difference averaged ± 0.3%.

Using volumetric bulk measurements, six intraplant simulated shipments of plutonium nitrate in a 10-liter container showed an average shipper-receiver difference of +0.04% and a relative standard of ± 0.25%.

Solution Composition

Within the composition range specified for AEC purchase of private plutonium nitrate (50-250 grams of plutonium per liter; 2-10 N nitric acid; less than 5000 ppm metallic impurities), no composition was found to cause a shipper-receiver bias. The physical and chemical measurement methods demonstrated in this program are applicable over this composition range.

Solution Stability

The stability of plutonium nitrate solutions is dependent upon the hexavalent plutonium content. Plutonium (VI) is a major constituent of plutonium nitrate solutions that have been concentrated by evaporation. Self-reduction of plutonium (VI) to plutonium (IV) occurs with the evolution of oxygen. The self-reduction proceeds with a half-life of approximately several weeks. In the self-reduction reaction, hydrogen ion is consumed in the ratio of approximately one hydrogen ion per plutonium (VI) ion. The acidity reaction is not sufficient to cause hydrolysis of plutonium (IV) if initial acidity is above 2 N.

Analytical Measurements

The major contributor to shipper-receiver differences in this work was the analytical determination of plutonium in solution. A suitable method has been demonstrated, but interplant comparisons have shown a significant analytical bias between two widely used and recommended methods.

10. Research Contract No. AT(29-1)-1106 $ 95,000

Title

Plutonium Oxide Shipping Measurements

Institution: Dow Chemical Company, Rocky Flats Plant

Principal Scientific Investigators: J. T. Byrne, C. E. Caldwell

Period of Contract: 1963 to 1965

Background & Principle

Plutonium oxide is a convenient form in which to ship plutonium between plant sites. It is one of the forms in which the Atomic Energy Commission will purchase privately-produced plutonium. The amount of plutonium shipped is expected to increase rapidly within the next several years. To minimize problems in buying and selling plutonium between the Atomic Energy Commission and private reactor owners, a thorough study of the shipper-receiver measurement problem was needed.

Within the AEC complex, there has been a history of differences between the shipper-determined and receiver-determined plutonium content of plutonium nitrate solutions. A comprehensive study aimed at reducing or eliminating this problem was recently completed. (See summary 11, above).

To preclude repeating this history of shipper-receiver differences with plutonium oxide, a program was initiated under the sponsorship of the USAEC Division of Nuclear Materials Management to eliminate, if possible, these differences.

Objectives

To determine the possible causes of plutonium oxide shipper-receiver differences, and to eliminate these differences if possible.

Procedure

A program was set up in which the administrator had complete control over both simulated-shipping and simulated-receiving functions.

In defining the problem, the probable causes for the shipper-receiver differences were placed in five categories: analytical bias, bulk measurement bias, sampling bias, loss during shipment, and clerical errors. These causes could also be arranged in six areas listed as follows:

1. Analytical method standardization error.
2. Plutonium oxide instability.
3. Plutonium oxide insolubility.
4. Bulk measurement calibration error.

5. Inadequate sampling procedures.
6. Incomplete cleanup of shipping containers.

Personnel at Rocky Flats were experienced in handling and measuring plutonium oxide on both an intraplant and an interplant basis. This experience, combined with information that could be obtained from other sites, was the basis for developing the current program for investigating the problems associated with the shipping of plutonium oxide.

In setting a goal of eliminating plutonium differences, it is necessary to qualify the term "eliminating". The intent of this study is to eliminate differences that are statistically significant at the 95 percent confidence level. The significance of a measured difference is determined by the number and standard deviation of the measurements. In the interplant shipments, 8 cans were shipped twice thus checking twice on the reliability of the measurements. With 8 measurements (seven degrees of freedom), a difference about equal to the standard deviation can be shown to be significant at the 95 percent confidence level. It was anticipated from previous experience with both plutonium nitrate and plutonium oxide that the measurement standard deviation (and consequently, the detectable bias) would be about 0.2 percent.

Conclusions

The major causes of shipper-receiver differences, during shipments of plutonium oxide, have been identified, and methods of minimizing the differences have been developed. Acceptable levels of shipper-receiver differences have been demonstrated, and recommendations for further improvement have been made. Plutonium oxide, properly prepared, is stable and sufficiently soluble to use as a shipping form. The equipment and procedures used in this study, for preparation of acceptable plutonium oxide, are recommended as specifications. Bulk-measurement errors are minimal when proper procedures and equipment are used. Retention of plutonium oxide in the shipping cans was found to be negligible. A template-sampling method appears to be adequate. A procedure for the non-destructive calorimetric determination of plutonium in a shipping container has been demonstrated.

The average shipper-receiver differences for 12 interplant shipments, of oxide prepared from oxalate or peroxide, between Hanford and Rocky Flats was +0.14 percent, with a standard deviation of 0.23 percent. Using analytical laboratory results, from the same laboratory only, the average shipper-receiver difference for these interplant shipments was +0.10 percent with a standard deviation of 0.18 percent. A similar group of 12 intraplant shipments, at Rocky Flats, gave an average shipper-receiver difference of -0.06 percent with a standard deviation of 0.19 percent.

The positive shipper-receiver difference (+0.14 percent) for the interplant shipments is explained by the fact that all receiving samples were analyzed in an atmosphere of lower humidity than that in which the oxide was initially prepared. Consequently, the analyses show a progressive increase in plutonium concentration because of loss of absorbed moisture. The interplant shipments also included two batches (four shipments) of oxide prepared by denitration of plutonium nitrate. Results from this material were erratic, indicating heterogeneity and instability because of incomplete calcination. The shipper-receiver differences obtained with denitrated oxide are not included in the above results.

The ceric sulfate redox titration method was used to determine plutonium for calculation of shipper-receiver differences. This method is subject to sampling error, and it requires that the oxide be completely soluble.

It is believed that the calorimetric method could be used for the determination of plutonium in shipping containers. The calorimetric method requires precise measurements of the isotopic composition. But it does not require: (1) that the oxide be homogeneous, (2) that it be completely soluble in a limited time, or (3) that careful weight measurements be made. The precision of the method is comparable to the ceric sulfate redox titration method as performed at Rocky Flats and Hanford (∼ 0.06 percent RSD). The bias between laboratories, for the calorimetric method, should be less than the bias found for the wet chemical methods.

11. Research Contract No. $ 8,600

Title

Plutonium Compound Evaluation Program

Institution: Five USAEC Laboratories

Principal Scientific Investigators:

Period of Contract: 1962 to 1966

Background & Principle

Three plutonium compounds, plutonium sulfate tetrahydrate, anhydrous plutonium sulfate and dicesium plutonium hexachloride, were evaluated for suitability as primary plutonium chemical standards to replace the plutonium metal standard. The use of a compound rather than metal would provide more versatility in use because of the ease of subdivision to take small portions.

A statistical evaluation of the data accumulated by the various laboratories which have been analyzing the three plutonium compounds gave no indication of decomposition trends for any of the compounds. The dicesium plutonium hexachloride salt showed the lower error mean square and appeared to be the preferred compound. Even when a laboratory had difficulty with the analyses there seemed to be less difficulty with the dicesium salt then with the other two compounds.

Looking at the compounds from a stoichiometric standpoint, none of the three are suitable for a primary standard. NBS requires that a primary standard be stoichiometric to within 0.02 parts per ten thousand while a working standard need be only to within 0.05 parts per ten thousand. Both the dicesium salt and the plutonium sulfate tetrahydrate would be suitable as working standards but neither are sufficiently stoichiometric to be a primary standard. The anhydrous sulfate salt is not suitable as either a primary or a working standard.

At a meeting held on March 2, 1966, the USAEC sponsored Advisory Committee for Standard Reference Materials and Methods of Measurement recommended that both the dicesium plutonium hexachloride and plutonium sulfate tetrahydrate be used as working standards. The Committee felt that both compounds should be made available to permit flexibility in chemical, isotopic and impurity analyses. Batches of these materials would be standardized against the primary standard metal and will be distributed either by NBS or NBL as working Standards.

12. Analyst Performance on Synthetic Purex Solutions (1/15/60)
WCAP-6036

H. Ginsburg, C. C. Thomas, Jr.

Results are presented on an evaluation of analyst performance and the reliability of certain analytical procedures employed in the Nuclear Materials Controls System (NMCS) program. Synthetic samples pertinent to the NMCS Project were prepared and analyzed. The bias, standard deviation; and their associated 95 percent confidence limits are presented for each analyst for several series of determinations. A modified X-ray fluorescence spectrometric procedure for the destructive assay of uranium in MTR and ETR fuel plates is discussed.

13. Research Contract No. AT(10-1)-1230 $ 383,000 to 9/30/68

Title

Qualification of Umpire Laboratories

Institution: Idaho Nuclear Corporation

Principal Scientific Investigator: J. E. Rein

Period of Contract: 4/29/65 to continuing

Background & Principle

Many private U. S. Companies and foreign concerns are entering the atomic energy industry. As a result, the AEC is transacting an increasing number of sales, leases, and purchases of fissionable material outside the AEC complex. The basis of the monetary payments or credits is the quantity and quality of the materials involved as established by analysis. Based on past experience, there will be inevitable differences and hence disputes between the AEC and other parties. Such disputes are best resolved by neutral umpire laboratories.

The types of materials which are currently being transferred within the AEC complex and which can be expected to be exchanged between the AEC and other parties include: uranium hexafluoride, uranyl nitrate (solution and crystal), uranium oxides (U_3O_8, UO_2, and UO_3), uranium metal, plutonium nitrate solution, plutonium dioxide, and plutonium metal. Of these, uranium hexafluoride, uranyl nitrate, and plutonium nitrate solution will be exchanged most frequently and highest priority has been assigned to them. The specifications of interest are various physical properties, impurities including nonradioactive metallic and nonmetallic elements, fission products, and certain actinide nuclides; and most important, concentration and isotopic assay of uranium and plutonium.

Objective

The purpose of this project is the establishment and administration of a program to qualify independent U. S. and foreign laboratories to serve as umpires for various uranium and plutonium materials. Laboratories will be qualified on the basis of the results of actual analyses of specially prepared standards.

Procedure

This program is being conducted in cooperation with other AEC-controlled laboratories including New Brunswick Laboratory (NBL), Oak Ridge Gaseous Diffusion Plant (K-25), Paducah Gaseous Diffusion Plant (PAD), Goodyear Atomic Corporation (GAT), Atlantic Richfield Hanford Co. (ARHCo), Rocky Flats (RF), and Los Alamos Scientific Laboratory (LASL).

The program is being implemented as follows:

Phase I: a. Review the specifications for the various uranium and plutonium materials.

b. Establish the availability of adequate methods for the specification analyses.

c. Establish the basic requirements of a prospective umpire laboratory.

d. Explain the objective of the Program to selected AEC laboratories and obtain their cooperation.

Phase 2: a. On the basis of information obtained in Phase I, inform laboratories seeking qualification what the Program entails, and to assess by means of a questionnaire, their interests, competences, and technical capabilities.

b. With the aid of the selected AEC laboratories, prepare standards of the various materials on the basis of information obtained in Phase I:a and analyze each standard for the various specifications.

Phase 3: a. Distribute the standards to those laboratories that qualify under Phase 2:a

b. Qualify or reject the laboratories on the basis of a personal inspection of their laboratory facilities and statistical comparisons of their data with that obtained under Phase 2:b by the AEC laboratories.

Status as of 4/30/68

A. Preparation and Analysis of Standards

1. Uranium Oxide

A second oxide has been prepared and tested for homogeneity by the analysis of six random samples for uranium and for iron. Uranium was determined because very precise methods are available and because it is the major component. Iron was determined because it is a major impurity constituent and because other investigators have indicated that its distribution in uranium oxide is a function of particle size. Both series of analyses indicated homogeneity.

In a supplemental study, the oxide was screened with a 200-mesh sieve and the two fractions were analyzed for iron. Contrary to the findings of other investigators, no significant difference was observed in the iron content of the two cuts.

2. Uranyl nitrate, U-233 enriched uranyl nitrate, and uranium hexafluoride

Standards of U-233-enriched uranyl nitrate were prepared at ICPP (Idaho Chemical Processing Plant of Idaho Nuclear Corporation) and standardized at K-25, GAT, NBL, and ICPP. All data that have been received from these laboratories have been tabulated for statistical evaluation.

The data received from the AEC standardizing and participating laboratories for the uranium concentration of uranyl nitrate, Standard A, has been statistically reevaluated. The reason for doing this is that the basic model was adjusted to allow qualification of laboratories and not analysis as was previously the case.

Test samples of this standard were sent to five U. S. and one foreign laboratory. The analysis results reported by laboratories seeking qualification on uranyl nitrate, uranium hexafluoride, and U-233-enriched uranyl nitrate were statistically evaluated to establish qualification. Several qualifying laboratories were visited to confirm capabilities.

3. Plutonium metal, plutonium nitrate, and plutonium oxide.

Standardizations of the plutonium nitrate, plutonium oxides, and plutonium metal standards were completed by ARHCo, LASL, NBL, and RF. Test samples of these materials were distributed to five U. S. and five foreign laboratories. Statistical evaluation of analytical results was started.

B. Preparation of Program Documents

The first of a series of documents for transmittal to the AEC Office of Safeguards and Materials Management has been prepared. This first document covers the basis of the Umpire Program. It will be followed by documents which will report analytical results, the statistical evaluation of these results, and will recommend laboratories for qualification.

Status as of 10/1/68

A. Preparation and Analysis of Standards

Uranium oxide samples have been shipped to eight participating organizations. Two of the commercial organizations have broadened their participation in this Program by indicating that, in addition to uranium oxide, they are interested in uranyl nitrate, plutonium nitrate, plutonium oxide and plutonium metal.

Statistical evaluation of data received from the participating laboratories as of September 30, 1968, for the concentration and isotopic distribution of UF_6 has been completed and the qualified laboratories determined. Three laboratories obtained acceptable results for the isotopic distribution of UF_6 for U-235 by surface ionization mass spectrometry techniques. The impurity categories for UF_6 are now in the process of being completed.

B. Preparation of Program Documents

The first two of a series of documents have been issued:

QUALIFICATION OF UMPIRE LABORATORIES FOR THE ANALYSIS OF URANIUM AND PLUTONIUM MATERIALS SPECIFICATIONS

Report I. BASIC PROGRAM

ABSTRACT

On behalf of the Office of Safeguards and Materials Management, USAEC, Idaho Nuclear Corporation is conducting a program directed toward the qualification of U.S. commerical and foreign government laboratories to serve as umpires in AEC transactions involving uranium and plutonium materials. Qualification is based on a demonstrated ability to analyze the various materials for the various specification categories. This first report describes the basic program. Supplementary reports will be issued which recommend the qualification of participating laboratories for each of the various materials.

QUALIFICATION OF UMPIRE LABORATORIES FOR THE ANALYSIS OF URANIUM AND PLUTONIUM MATERIALS SPECIFICATIONS

Report II. First Report on Uranyl Nitrate

ABSTRACT

As indicated in "Report I. BASIC PROGRAM", issued May, 1968, the Idaho Nuclear Corporation is conducting a program, on behalf of the Office of Safeguards and Materials Management, directed to the qualification of U. S. commercial and foreign government laboratories to serve as umpires in AEC transactions involving uranium and plutonium materials Qualification is based on a demonstrated ability to analyze the various materials for the various specification categories. This report lists the specifications and describes the preparation and standardization of the uranyl nitrate standards. The statistical analyses and qualification recommendations are presented for those participating laboratories that have completed the analysis of this material at this time.

The third report, "Report III. First Report on Uranium Hexafluoride", is now being prepared. The contents of this report will be similar to that of "Report II."

Following "Report III", the proposed order of report preparation is plutonium nitrate, plutonium oxide, plutonium metal, U-233 enriched uranyl nitrate and uranium oxide.

14. Research Contract No. $

Title

Isotopic Composition Analysis by X-ray Fluorescence

Institution: Institute fur Radiochemie, Karlsruhe

Principal Scientific Investigator: A. von Baeckmann

Period of Contract:

Objective

Determine the accuracy, time required and cost of measuring isotopic composition. Investigate the possibility of using this method in an industrial plant for highly active uranium and plutonium containing solutions.

15. Research Contract No. $

Title

Isotopic Composition Analysis by Isotopic Dilution Spectrometry

Institution: Institute für Radiochemie, Karlsruhe

Principal Scientific Investigator: A. von Baeckmann

Period of Contract: August 1967 to (continuing)

Objective

Determine the accuracy, time required and cost of measuring isotopic composition and isotopic content by dilution spectrometry. The possibility of using this method in an industrial plant will be evaluated.

16. Research Contract No. $

Title

Isotopic Composition Analysis by Photometry

Institution: Institute für Heisse Chemie, Karlsruhe

Principal Scientific Investigator: A. von Baeckmann

Period of Contract: August 1967 to (continuing)

Objective

Determine the accuracy, time required and cost of measuring isotopic composition by photometry. The study will include an evaluation of the possibility of using this method in an industrial plant.

17. Research Contract No. AT-33-1-GEN-53 (Related to Safeguards) $ 20,000 to 6/30/68

Title

Rapid Process Control of Alpha Emitting Isotopes

Institution: Mound Laboratory, Miamisburg, Ohio

Principal Scientific Investigators: R. J. Baltisberger, W. R. Wood, Jr.

Period of Contract: 7/1/67 to 6/30/68

Background & Principle

A probe type alpha detector was developed to assay polonium content in process solutions. The desirable features of the probe are its adaptability to in-line installation and the rapid results read directly from an electrometer amplifier. These features allow better production control.

The detector is an air-filled ionization chamber which operates on 200 V and responds almost exclusively to alpha disintegrations. The response of the electrometer varies nearly directly with the alpha disintegrations of the measured solutions.

Experimental analyses were conducted exclusively with HCl solutions of polonium with varying bismuth concentrations simulating actual process solutions. The instrument was effective in measuring polonium alpha activity in HCl acid solutions between 0.008 and 0.8 mCi/ml. Accuracies of ± 10% were obtained routinely.

Published reports:

"Simplified Alpha Probe Type Detector" Paper to be presented at Twelfth Annual Analytical Conference, Gatlinburg, Tennessee October 8-10, 1968 by R. J. Baltisberger and W. R. Wood, Jr. Paper is being submitted for publication in Analytical Chemistry.

B. Calculation of Heavy Isotope Content of Reactor Fuel

	Page
1. Determination of the Tritium Build-up in a Heavy-Water Moderated Reactor (IAEA) 129/RB	37
2. Analytical and Experimental Methods of Determining Heavy Isotope Content of Operating Fuel Elements (USAEC)	39
3. Theoretical Analysis of Plutonium Buildup and Uranium Depletion in Power Reactors (IAEA) 467/CF	43
4. Theoretical Analysis of Plutonium Buildup and Uranium Depletion in Power Reactors (IAEA) 468/RB	45
5. Theoretical Analysis of Plutonium Buildup and Uranium Depletion in Power Reactors (IAEA) 495/RB	48
6. Theoretical Analysis of Plutonium Buildup and Uranium Depletion in Pressurized Water and Boiling Water Power Reactors (IAEA) 513/RB	49

1. Research Contract No. 129 $ 9,320

Title

Determination of the Tritium Build-up in a Heavy-Water-Moderated Reactor

Institution: Institute of Nuclear Sciences "Boris Kidric"
Belgrade, Yugoslavia

Principal Scientific Investigators: Z. Vukmirovic, S. V. Ribnikar

Period of Contract: 12/27/61 to 12/26/63

Background & Principle

Heavy hydrogen, H^2, shows a relatively small thermal neutron activation cross section of 0.57 mb. However, in a heavy-water-moderated nuclear reactor, high neutron fluxes after some time produce quite appreciable quantities of tritium, H^3. The amount of tritium, after decay corrections, will be proportional to the entire neutron dose received by the heavy water, as well as to the integral energy released by the reactor fuel.

The only uncertain point is the migration behavior of the tritium through the reactor system. A part of the tritium might escape from the heavy water after its formation with the deuterium gas produced by radiolysis of the heavy water. False results could therefore be obtained for the above-mentioned activity-energy relationship.

The intention of the present investigation was to prove the reliability of a correlation between the amount of tritium found in the reactor heavy water, and the power history of the reactor.

Procedure

The investigation was made on the 6.5 MWt heavy water moderated reactor (2.0% U-235 enrichment) at Vinca, Yugoslavia.

Samples of both the heavy water from the moderator tank and the hydrogen liberated by the hydrolysis were taken over a 24-month period. The H^3 activity was determined using liquid scintillation counting or internal gas counting.

Results

A correlation between the reactor operation data obtained by thermal measurements and the corresponding values obtained from the tritium activity measurements was obtained. A previously known conversion factor, the average integral neutron flux per unit power, 0.41×10^{13} n/cm^2/sec per megawatt was used.

The integrated power of the reactor was determined by thermal and tritium activity measurements agreed well within the measurement error limits of the thermal data (± 10%) and the activity data (± 5%).

Conclusions

The tritium activity in heavy water moderator corresponds to the power history of a reactor and can be used for measurements of the integral neutron flux and integral power release of it. The average integral neutron flux per unit power has to be known for each particular reactor.

2. Research Contract No. AT(38-1)-428 $ 217,000 to 1/29/68

Title

Analytical and Experimental Methods of Determining Heavy-Isotope Content of Operating Fuel Elements (9/30/65)

Institution: Combustion Engineering, Inc.

Principal Scientific Investigators: M. F. Valerino, Z. R. Rosztoczy

Period of Contract: 1964 to February 1968

Background & Principle

For the purpose of determining the monetary value of fissile material in irradiated fuel elements, it is necessary that the content of uranium and plutonium be calculated. Since fuel discharged from a power reactor is generally not processed for several months, an interim monetary settlement must be based upon calculations. A knowledge of the nuclear material content of discharged reactor fuel is also required in the implementation of safeguards procedures to minimize or detect the clandestine diversion of such material.

This study, conducted by Combustion Engineering, Inc. since 1964 for the Division of Nuclear Materials Management, is covered by a three-phase contract.

Phase I

The specific objectives were to determine the capabilities and limitations of the present calculation methods, to evaluate the accuracy of the available techniques, and to express as quantitatively as possible the agreement which might be expected between calculational results and the results of a careful dissolution chemical analysis.

The current calculational techniques used to predict heavy-isotope content of nuclear fuels exposed in power reactors were identified and evaluated from the standpoint of estimating accuracies of predictions. A significant number of experimental measurements were made for comparison with the calculated heavy-isotope content of irradiated fuel.

Phase II

Work under this phase showed that a GE-developed code, FLARE, could be used by the AEC to verify the results of more detailed calculations as reported by the reactor operations. FLARE is a burnup distribution code, it apportions total burnup (as determined from heat balance calculations) among the fuel assemblies of the reactor. In most cases it will be possible to make one mesh point in the calculation correspond to a single fuel assembly in the reactor.

The FLARE code provides the best compromise for calculating relative fuel assembly exposures which compare very favorably with experimental distributions inferred from flux wire, isotopic composition and Cs-137 measurements. The exposure predictions are most accurate for pressurized water and boiling water reactors which are predominant in current commercial power production and for which significant experimental and operational data are available. Data were reviewed in detail for, among others, the following major power reactors: Indian Point, Dresden, Shippingport, Hallam, Saxton, Carolinas-Virginia, Piqua and Yankee. The same degree of accuracy is expected for other reactor types having thermal or near thermal neutron spectra.

Phase III

Phase III covers the development of a method to independently check the heavy isotope contents of operating fuel elements. Detailed procedures were developed to apply the FLARE code to each of 17 U. S. power reactors.

Research Contract No. AT(38-1)-428 (continued) $ 25,000
in FY-68

Title

Analytical and Experimental Methods of Determining Heavy Isotope Content of Operating Fuel Elements

Institution: Combustion Engineering, Inc.

Principal Scientific Investigators: R. Kern, A. T. Shesler

Period of Contract: 7/1/67 to 6/30/68

Background & Principle

Under previous contracts supported by the USAEC, Combustion Engineering, Inc., has developed a procedure designated ISOCHECK for calculating U-235 burnup and Pu production in power reactors. ISOCHECK is not a computer program but is a method or procedure of applying existing computer programs (FORM, TEMPEST, SCEPTER, etc.) to develop some of the input data necessary to adapt the general burnup program (FLARE) to an individual reactor. ISOCHECK combines FLARE and another code, CETIC, to permit calculation of the burnup and production. FLARE is a computer code which calculates power distribution from reactor physics and design data. CETIC converts the exposure distribution into a core inventory of nuclear material by combining FLARE output and reactor operating data. It should be observed that ISOCHECK represents a compromise between accuracy of results and simplicity of calculations for determining the heavy-isotope content in the operating fuel elements of power reactors. ISOCHECK must be modified (or updated) if a different fuel element is used (cladding, enrichment).

ISOCHECK has been applied to 17 power reactors in the United States. However, because of design changes to the Yankee, Dresden I and Humboldt reactors, ISOCHECK is no longer valid.

Although ISOCHECK has worked very well on existing reactors, it appears that it may not give as accurate results when applied to the much larger reactor cores now being designed. It is desirable to determine if and how the procedure can be modified for such application.

Objectives

1. Update ISOCHECK for application to Yankee, Dresden I and Humboldt reactors.
2. Conduct a feasibility study to determine how the FLARE code can be expanded for use on large reactors.

Procedure

As stated above

Status as of 6/1/68

1. Formal reports covering the updating of ISOCHECK for the three reactors have been completed.
2. The feasibility study has been completed.

3. Research Contract No. 467 $ Cost-Free

Title

Theoretical Analysis of Plutonium Buildup and Uranium Depletion in Power Reactors

Institution: Atomic Energy of Canada Ltd., Chalk River, Ontario

Principal Scientific Investigators: M. F. Duret, M. J. Hansall

Period of Contract: 10/1/66 to 3/31/67

Background & Principle

Calculations are required on the plutonium buildup and uranium depletion in the natural uranium heavy water moderated and cooled CANDU type reactors, namely, NPD, Douglas Point and Pickering. The work will be part of a coordinated programme of research in theoretical analysis of plutonium buildup and uranium depletion in power reactors.

Procedure

Two computer codes were used to perform the calculations:

(i) LATREP (AECL-2548), which calculates lattice parameters for any cell geometry and lattice pitch. LATREP was used to evaluate initial Westcott r values, neutron temperatures, fast fission ratios and initial values of the effective ^{238}U capture cross-section. Resonance capture in the LATREP cases was multiplied by 0.9, a factor which improves agreement with measured relative conversion ratios.

(ii) CARIBOU (AECL-2296) which uses LATREP spectrum parameters to calculate isotope reaction rates. The burnup equations are integrated and an interpolation routine evaluates the isotopic composition of the fuel for specified burnups in MWd/te of natural uranium. Isotopic compositions are given in kg/te, or gm/kg, of natural uranium.

Conclusions

The isotopic composition of irradiated natural uranium fuel was calculated for the three reactors NPD, Douglas Point (CANDU-PHW) and Pickering, and also for a conceptual 18-rod fuel design in CANDU-BLW. Three sets of results were prepared for each reactor. These correspond to operation at the design lattice pitch and also at pitches chosen to reduce and increase the moderator volume per cell by 20%.

The results are tabulated in two forms:

44

(i) at burnup intervals of 500 MWd/te up to 10,000 MWd/te.

(ii) at irradiation intervals of 0.1 n/kb to 4 n/kb.

The results of fuel composition versus burnup are also given graphically, but only for the normal lattice pitch, as differences with changing pitch tend to be rather small.

Comparison with Experiment

Precise measurements were made of the composition of an NPD bundle irradiated to about 5600 MWd/te. The measured U-235 concentration relative to its initial value in this bundle was 0.3905. This value has been used for interpolating in the CARIBOU tables of isotopic composition. The calculated Pu-241 concentration has been reduced by 3.5% to allow for decay in the 9 months between removal of the bundle from the reactor and the analysis of the bundle. The final results are tabulated below, the plutonium composition being given in atom percentages.

	EXPERIMENT	CARIBOU	CARIBOU/EXPERIMENT
^{235}U	.3905 ± .0009	.3905	
Pu/U (atom ratio)	.003133 ± .000020	.003136	1.001
^{239}Pu (atom percent)	73.34 ± .06	73.49	1.002
^{240}Pu (atom percent)	22.04 ± .06	21.94	.995
^{241}Pu (atom percent)	3.815 ± .01	3.783	.992
^{242}Pu (atom percent)	0.812 ± .002	0.790	.973

4. Research Contract No. 468 $ 7,500

Title

Theoretical Analysis of Plutonium Buildup and Uranium Depletion in Power Reactors

Institution: United Kingdom Atomic Energy Authority,
Risley, Lancashire

Principal Scientific Investigator: B. Cutts

Period of Contract: 10/1/66 to 9/30/67

Background & Principle

Calculations on plutonium buildup and uranium depletion covering the natural uranium graphite-moderated gas-cooled reactors of Magnox type are required. A great deal of information based upon old calculation methods is already available. This will be updated by using new methods and computer codes.

Plutonium production is affected by geometry, temperature and nuclear characteristics of the graphite, fuel, canning and inter-fuel materials. To cover even a relatively small number of these factors and their interactions, at (say) three levels, would have resulted in an extremely large number of cases. It was therefore decided to reduce the investigation to a practical size by restricting the study to the following variables which are parametric in the calculation:

(a) Moderator/fuel volume ratio

(b) Resonance integral of U-238

(c) Moderator temperature

(d) Mean fuel rating

To assess the range of variation to be covered in the investigation, a general survey of the design features of existing UK magnox reactors was carried out. The parameters varied to some extent within the reactor cores and with the operating condition of the reactor but a representative range for normal conditions of these reactors was chosen for this study.

Procedure

Plutonium Production Calculations

The calculations of plutonium production were carried out using the method of lattice calculations known as ARGOSY. In this method, the mean fuel and moderator data for the lattice cell are evaluated in three energy groups. Below 4 eV a generalised heavy gas thermalisation model is assumed, together with a collision probability technique for transport effects. In the resonance region between 4 eV and 10 keV an effective resonance integral of U-238 is used which is consistent with Hellstrand's values. Above 10 keV a few-group collision probability method is employed. The effect of cell environment is allowed for through energy dependent bucklings.

A comparison of predictions of plutonium build-up using ARGOSY and mass spectrometer measurements made on samples taken from fuel irradiated in the Calder Hall and Windscale A.G.R. reactors has been completed. Although it was shown that the agreement between theory and measurements is quite good, the author suggested modifications to the ARGOSY library data which would improve the agreement; these have been incorporated in the present study. It should be noted that the modifications are introduced only for the purpose of calculating plutonium production.

Burn-up Calculations

The calculations were carried out using a typical C.E.G.B. Bradwell reactor lattice cell as the basic lattice, and perturbing it to give the required values of the parameters.

In the ARGOSY method it is necessary to input thermal flux hyperfine weighting factors for the can and end cap materials. These were assessed by representing the fuel element in RZ geometry with the end cap material smeared out over the appropriate region, and carrying out a thermal group diffusion theory calculation with zero flux gradient boundary conditions at the edges of the cell. The calculation indicated flux weighting factors relative to the fuel of 1.20 for the cans and 1.69 for the end cap materials. The latter value seems to be high and would be reduced if the gap materials had been more localised in the calculation. However, trial ARGOSY calculations with values of the flux weighting factor in the end cap region of 1.69 and 1.20 showed negligible difference in the total build-up of plutonium at 5000 MWd/teU. This calculation illustrates that plutonium production is quite insensitive to large variations in the amounts of magnox material present in the fuel cell even though the reactivity might be adversely affected.

In a reactor situation the presence of vacant lattice positions in the core can result in the reactor spectrum being significantly different from that of the full lattice spectrum. With the ARGOSY three-group scheme of lattice cell calculation, some allowance for this effect is possible by a technique in which "fictitious" bucklings are input to produce the change to the lattice spectrum. To determine an estimate for the bucklings, an ARGOSY burn-up calculation was performed on the Bradwell lattice cell and the three-group nuclear parameters were time-averaged over an irradiation range from 0 to 3500 MWd/teU. A 5 x 5 supercell calculation was then carried out with a central vacancy surrounded by the 24 channels having the time-averaged properties and the average three-group fluxes in the surrounding cells determined. The 5 x 5 array was selected as typical of the control densities expected, although Bradwell is actually 1 in 18. The average buckling in the cells surrounding the vacancy were then obtained for each group by solving the appropriate equations. These calculated bucklings were then used in the final set of ARGOSY calculations.

The ARGOSY output, relevant to the present study, comprised the number densities of uranium and plutonium isotopes in the irradiated fuel at approximately 200 MWd/teU intervals up to 5200 MWd/teU. A data processing programme was developed for the IBM 7090 which used a quadratic interpolation to give the number densities at exactly 200 MWd/teU intervals and then calculated the quantities required in accordance with the appropriate equations. The use of 3rd and 4th order interpolation was investigated and found to have no significant effect on the calculated values. The results for each case were then printed out in tabular form ready for photographic reproduction.

Conclusions

The total plutonium and Pu-240 concentrations at 5000 MWd/teU, expressed as a percentage of the total plutonium, have been averaged to illustrate the main effects of the parameters considered in this study. It was observed that changes in moderator/fuel volume ratio and moderator temperature have the largest effects on total plutonium and % Pu-240 and that even these over the range considered, do not alter the total plutonium concentrations by more than 6%. The effects of altering the resonance integral and rating are not more than 1%. When considering variations of the order of 1%, it is worth noting that some of the assumptions necessary to specify the problem yield effects of this magnitude; for example, the effect of including fictitious bucklings in a typical ARGOSY calculation is to increase the total plutonium at 5000 MWd/teU from 2.872 to 2.901 kg/teU, an increase of about 1%.

5. Research Contract No. 495 $ 30,000

Title

Theoretical Analysis of Plutonium Build-up and Uranium Depletion in Pressurized Water and Boiling Water Power Reactors

Institution: I. V. Kurchatov Atomic Energy Institute, Moscow, USSR

Principal Scientific Investigator: Professor S. U. Feinberg

Period of Contract: 1/1/67 to 6/30/68

Scope of Contract

The study is intended to provide basic material for further investigations on plutonium recycling as well as for practical application which is of interest to the Division of Safeguards and Inspection as well as the Division of Nuclear Power and Reactors. The calculations on plutonium burnup and uranium depletion will cover pressurized water power reactors and boiling water power reactors, for varying basic reactor parameters such as fuel enrichments, type of fuel cladding, size of fuel elements etc., in a wide range of fuel burnups employing the techniques, methods and information developed in the Soviet Union.

The study of PWR and BWR fuel burnup and plutonium buildup will include a large number of gases covering many variables. The sequence of work is expected to be as follows:

1. assembling available source data on reactor parameters and nuclear data;
2. developing the computer programme for this specific requirement;
3. calculations on the computer;
4. checking the calculations against other available data;
5. presenting the results in the form of graphs and tables required for the safeguards activities;
6. preparing the report on the project.

The total financial support desired from the Agency for the two years' project will be $30,000. This would represent only a small part of the total expenditure for carrying out the work.

6. Research Contract No. 513 $ 31,000

Title

Theoretical Analysis of Plutonium Buildup and Uranium Depletion in Pressurized Water and Boiling Water Power Reactors

Institution: Battelle Memorial Institute, PNL, Richland, Washington, U. S. A.

Principal Scientific Investigators: E. A. Eschbach, D. E. Deonigi

Period of Contract: 6/15/67 to 6/14/68

Background & Principle

Calculations are required regarding the buildup of plutonium and the depletion of uranium in typical reactors in the United States.

Procedure

Typical pressurized water reactor (PWR) and boiling water reactor (BWR) designs selected for study are, respectively, Diablo Canyon and Browns Ferry. Uranium and plutonium isotopic depletion and yield data for each of these designs were calculated by ZODIAC code for operation at 1000 MWe. Additional calculations to evaluate effects of UO_2 fuel rod diameter and of moderator/fuel volume ratio variations were also made. Comparison of ZODIAC-derived data with available analytical data, including consideration of changes in operating conditions for exposures first to about 14,000 and subsequently to about 24,000 MWD/Te in a PWR, indicates good correlation. Analytical data for BWR isotopic depletion and yields are not yet available, with adequate definition of operating conditions leading to compositions reported, to permit a detailed comparison with calculated data as for PWR. Comparisons between PWR and BWR calculated data are expected, however, to lead to valid conclusions concerning isotopic composition and yield differences anticipated under operating conditions.

ZODIAC calculations, if carried out for each of the 360 cases forming the subject of this study, would require not less than 180 on-line hours of computer time. Battelle-Northwest's ALTHAEA code, calibrated to provide calculated data equivalent to those derived by ZODIAC, will be employed rather than ZODIAC for the bulk of the calculations. Costs for ALTHAEA machine time are only a small fraction of similar costs for ZODIAC. Code calibration, consisting essentially of adjustment of variable code input values to provide calculated data equivalent to those obtained from other sources such as ZODIAC or analytically determined values, has been completed.

Computer code calibration studies, referred to above, have been performed for cases using zircaloy-2 clad enriched UO_2 fuel for the reference PWR and BWR designs. Several ALTHAEA code calculations, at conditions identical with zircaloy-clad cases except for the substitution of stainless steel as cladding material, have also been completed. The magnitude and direction of changes in isotopic depletion and yields resulting from the introduction of additional parasitic neutron absorptive material is illustrated by these data.

Conclusions

The desired calculations are being completed, based upon PWR and BWR design data and certain simplifying assumptions, for selected cases of stainless-steel clad and zircaloy-clad fuels.

C. Fuel Element Analysis

Page

Non-Destructive

1. The Development of a Method of Nondestructive Analysis to Determine the U-235 and Pu Content of Irradiated Fuel Elements That Are in Storage Under Water and That Have Been Out of the Reactor for Periods of More Than One Month (IAEA) 4/RB 53

2. Nondestructive Method for Evaluating the U-235 and Pu Content of Irradiated Fuel Elements (IAEA) 5/RB 54

3. Research on the Nondestructive Analysis of Irradiated and Unirradiated Fuel Elements for Nuclear Reactors (IAEA) 7/RB 56

4. Nondestructive Analysis of Irradiated Fuel Elements Using a Flux Integrating Monitor (IAEA) 41/RB 57

5. Nondestructive Analysis of Irradiated Fuel Elements by Gamma Ray Scanning (IAEA) 47/RB 59

6. Development of a Method of Nondestructive Analysis of Irradiated Fuel Elements for Uranium-235 and Plutonium Content by Monitoring and Spectrometry (IAEA) 87/RB 61

7. A Feasibility Study of Nondestructive Assay of Plutonium-239 in Irradiated Fuel Rods Using Slowing Down Time Spectrometer (IAEA) 92/RB 64

8. Nondestructive Analysis of Irradiated Fuel Elements by Establishing a Reliable Correlation Between One or More Gamma Lines From the Fission Products and the Burn-up of Irradiated Fuel Elements or Samples (IAEA) 163/RB 66

9. Nondestructive Testing of Fuel Elements by Means of Compton Recoil Spectrometry (IAEA) 180/RB 69

10. A Gamma-Ray Scanner for Safeguards Use (USAEC) 71

11. The Fuel Assay Scanner (USAEC) 72

12. Field Testing of the Fuel Assay Scanner (USAEC) 73

13. A Method for the Nondestructive Analysis of Power Reactor Fuel Elements (USAEC) 74

14. Nondestructive Assay of Nuclear Fuel (USAEC) 75

15. Nondestructive Analysis of Burnup in Irradiated Fuel Elements (USAEC) 77

16. Pulsed Neutron-Research for Nuclear Safeguards (USAEC) 78

17. Investigation of Photoinduced Reactions for Non-destructive Nuclear Materials Safeguards Applications 90

18. Laser Microprobe Analyser (USAEC) 96

19. High-Resolution Gamma Spectrometry for Analysis of Irradiated Fuel (USAEC) 98

20. Isotopic Composition by Delayed Neutrons-Unirradiated Fuel (BMwF) 101

21 Gamma-Ray Assay of Pu-238 in Waste Drums (USAEC) 102

22. In-Line Gamma-Ray Assay of Pu-238 in Waste Cans (USAEC) 103

23. Safeguards Applications of Precision Mass Spectrometry (ACDA) 105

24. Plutonium and Uranium Content in Fresh Fuel Elements Using Slowing-Down Time Techniques (BMwF) 107

25. Plutonium and Uranium Content in Unirradiated Fuel Pins (BMwF) 108

Destructive

26. Destructive Analysis of Unirradiated and Irradiated Fuels (IAEA) 121/RB 109

27. Service Contract--Analysis of Irradiated Fuel Plate from BGRR (Brookhaven Graphite Research Reactor) (IAEA) 314/RB 110

1. Research Contract No. 4 $ 25,000

Title

The Development of a Method of Nondestructive Analysis to Determine the U-235 and Pu Content of Irradiated Fuel Elements That Are in Storage Under Water and That Have Been Out of the Reactor for Periods of More Than One Month

Institution: Commissariat a l'Energie Atomique , France

Principal Scientific Investigators: V. Raievski, B. Sautiez

Period of Contract: 1/17/59 to 12/31/60

Background & Principle

Measuring the plutonium content of irradiated fuel elements is one of the problems of accounting of fissionable materials. The method of measurement, if based on fission induced by thermal neutrons, must rely on a significant physical difference between the U-235 and Pu nuclei. Here a method based on the difference of probabilities for the emission of certain numbers of neutrons during fission by U-235 and Pu nuclei was examined.

Procedure

Measurements were made with uranium and plutonium samples. The results indicated that the method could clearly distinguish between U-235 and Pu but, with the technique applied, the method could not determine the actual quantity of Pu with any acceptable accuracy. A number of improvements were suggested.

Conclusions

Further efforts would be required to improve the method before it could be applied to routine testing of fuel elements.

2. Research Contract No. 5 $ 8,000

Title

Nondestructive Method for Evaluating the U-235 and Pu Content of Irradiated Fuel Elements

Institution: Centre d'Etude de l'Energie Nucleaire
Mol, Belgium

Principal Scientific Investigators: J. Juliens, F. Motte, J. Bai

Period of Contract: 12/20/58 to 12/31/60

Background & Principle

The proposed method is a contribution to the solution of this complex problem; it consists of determining the concentration of U-235 and Pu-239 atoms by counting the prompt fast neutrons and the delayed fast neutrons emitted by the element when exposed to a beam of slow, low-intensity neutrons.

The prompt and delayed "secondary" neutrons emitted by the element being analyzed can be discriminated from the slow "primary" neutrons bombarding the element on the basis of their differences in energy. The proposed method involves various calibrations.

Procedure

A beam of low energy neutrons, i.e. thermal neutrons contaminated by intermediate neutrons, was emitted by the BR-1 reactor and bombarded the fuel element being analyzed. The slow flux at the site of irradiation was approximately 2 X 10^5 n/cm^2/s.

The fast neutrons, both prompt and delayed, were detected by scintillation in crystals containing charges of B and ZnS(Ag) mounted on photomultiplier tubes.

Conclusions

The results obtained appeared to be within 7% of the true uranium content of the fuel element tested.

The resulting accuracy could be very easily increased by using a beam of higher intensity or by increasing the geometrical efficiency of the counting device, which in these tests was very low as only one crystal was used.

This method has its attractive aspects and also its drawbacks. Although the tests were carried out with inactive elements only, the method could, it would appear, be adapted to the analysis of irradiated elements without the high activity of the fission products constituting a major obstacle.

On the other hand, a possibly serious remaining problem is the influence on the results of the distribution of fissionable nuclei inside the irradiated fuel elements.

3. Research Contract No. 7 $ 3,500

Title

Research on The Nondestructive Analysis of Irradiated and Unirradiated Fuel Elements For Nuclear Reactors

Institution: USAEC, subcontracted to Westinghouse, U. S. A.

Principal Scientific Investigators: S. L. Ruby, D. G. Gardner

Period of Contract: 3/19/59 to 6/18/59

Background & Principle

At the request of the Agency, the USAEC undertook a conceptual study to make a review, study and analysis of available unclassified information on the subject of nondestructive analysis of irradiated and unirradiated fuel elements for nuclear reactors, and submit to the IAEA a report based thereon containing the following information:

1. a review of the results of studies already undertaken in this field;
2. a determination of the areas that appear to be most promising for future research;
3. recommendations as to the best lines for future research to be undertaken.

Procedure

At the time the Agency invited the USAEC to undertake this project, the Westinghouse Electric Corporation was actively engaged in the large Nuclear Materials Control System study for the Atomic Energy Commission. Since a professional staff was already working on such a closely related study, it was deemed that this project could be completed most efficiently and economically by an amendment to the existing contract. Accordingly, Westinghouse Electric Corporation was selected as sub-contractor to conduct this study.

Conclusions

This study resulted in the preparation of a report by Westinghouse, WCAP-6014, titled "Survey and Selected Bibliography of Reactor Fuel Element Assay Systems" (6/10/59). This report contains a selected bibliography of 109 references on the nondestructive assay of irradiated and unirradiated reactor fuel assemblies. The report also includes a discussion and evaluation of these papers in the light of the problems presented by variations in enrichment, alloying, cladding, and geometrical arrangement, among the many kinds of fuel assemblies. A description of the theory of each method, the equipment utilized, and estimates of the precision obtainable are presented.

4. Research Contract No. 41 $ 5,200

Title

Nondestructive Analysis of Irradiated Fuel Elements Using a Flux Integrating Monitor

Institution: Institute of Nuclear Research
Warsaw, Poland

Principal Scientific Investigator: J. Aleksandrowicz

Period of Contract: 12/14/59 to 7/14/61

Background & Principle

The purpose of this contract was to determine burn-up in a two megawatt water cooled and moderated research reactor using cobalt wires attached to the reactor fuel elements. Fuel elements (10% enrichment) are made of U-Mg-ceramics, cylindrical in shape and Al clad. Burn-ups of 1.2 to 4.4% were investigated.

Procedure

The total burn-up in a fuel element was determined by integrating the burn-up in a cross section of the fuel element throughout its length. The detector selected was Co-59 in the form of a special Co-Al alloy. After irradiation in the reactor the activity of each centimeter of wire was measured by means of a scintillation counter. The integration of the activity distribution along the fuel rod was proportional to the burn-up in that rod during the time that the wire was attached to it.

The purely theoretical correlation between the activity of the monitor and the burn-up is complicated by the following two facts among others:

(a) The neutron absorption cross section of Co^{59} has two strong resonances at 135 eV and at 5000 eV;

(b) The neutron spectrum (especially the resonance spectrum) is different in the periphery of the fuel elements and within them.

For this reason a direct calibration of the method was necessary.

Conclusions

The need for calibrating the method by way of a special test loop for each type of fuel element and reactor makes its application difficult. The technical problems associated with attaching and detaching the wires to and from the fuel elements are serious.

The main source of error appeared to be in the detector itself, i.e. lack of uniformity in the diameter of the wire and/or in the distribution of Co along the wire.

The final answer regarding the possible accuracy of this method would be given by a chemical analysis of one or more of the monitored fuel elements. It has not been possible so far to perform such an analysis.

5. Research Contract No. 47 $ 16,000

Title

Nondestructive Analysis of Irradiated Fuel Elements by Gamma Ray Scanning

Institution: Institutt for Atomenergi
Norway

Principal Scientific Investigators: P. Kristiansen, T. Rogeberg

Period of Contract: 12/16/59 to 12/31/61

Background & Principle

In response to a request by the IAEA, the Institutt for Atomenergi submitted a proposal to the Agency to study the use of gamma-scintillation spectroscopy as a means of identifying and measuring the concentration of one or more fission products in irradiated fuel elements. If the concentration of the fission products could be determined, one would in principle have a measure of the burn-up of the fissile material.

During the discussions on the above proposal between representatives from IAEA and IFA, a variant of the proposed method was suggested by the IAEA representative. Excitation of characteristic X-rays from uranium and plutonium by β- and γ-rays from fission products should offer the possibility of measuring the concentration ratio of the heavy elements in irradiated fuel elements.

It was decided to examine both methods:

Method I:

By use of a crystal spectrometer, an attempt has been performed to measure the intensity of the K_α lines from Pu, relative to those of U. By this method, one aims at measuring the Pu content of the fuel.

Method 2:

By use of a scintillation spectrometer, the gamma spectrum from fuel samples has been measured. From this, one has tried to determine the content of the isotope Cs-137, and, thereby, the burn-up of U-235.

Procedures and Conclusions

Method 1:

Using test sample irradiated fuel slugs one was able to resolve and identify the Ka_1 lines in the second order spectrum from Pu and U. Measuring on test slug 1, the UKa_1 peak was visable;

the counting rate, however, was very low. No Pu peaks appeared. Measuring on test slug 2, the UKa_1 peak was clearly visable, and a small peak appeared which partly may be due to the Pu Ka_1 X-rays. The composition of the peak will have to be established by further experiments involving higher Pu concentrations.

If the peak is solely due to Pu Ka_1 emission this would indicate a very great concentration of Pu in the outer layers of the rod.

No estimate of the Pu content could be performed from the measurements, and the method seems not suitable for measuring such low Pu concentrations as occurred in the available test slugs.

The method is expected to be more successful when higher Pu concentrations are involved.

Method 2:

The first main problem was to separate the 0.661 MeV Cs-137 line from the 0.724, 0.759 and 0.764 MeV activities due to Zr-95 - Nb-95 decay.

The second main problem was to relate the measured Cs activity to the Cs-137 content of the rod, taking into account attenuation in the collimators, radial distribution of fission products and other geometrical corrections.

The final value for the Cs content agrees with the results of the chemical analysis within 25%.

Allowing for a possible 10% error in the quoted data for the fission yield of Cs-137, it seems that the burn-up of U-235 has been determined within an error of 35%.

It is believed that by improving the experimental conditions as to background, etc., the Cs content could probably be determined with an accuracy of 10 - 15%.

6. Research Contract No. 87 $ 22,000

Title

Development of a Method of Nondestructive Analysis of Irradiated Fuel Elements for Uranium-235 and Plutonium Content by Monitoring and Spectrometry

Institution: SNAM-Laboratori Riuniti Studi E Ricerche
San Donato Milanese, Italy

Principal Scientific Investigators: R. Renzoni, G. Cristiani

Period of Contract: 12/15/60 to 11/30/63

Background & Principle

One of the most important variables for the operation of nuclear reactors is the U-235 fraction burnt-out in the fuel element. Secondly, it is important to know the amount of plutonium produced (especially in natural uranium fuel elements where the plutonium value is particularly relevant from an economic point of view).

Unfortunately, no method of analysis of the above isotopes in fuel elements has been found up to now that would combine precision, speed of measurement and low cost.

The analysis is usually performed destructively, i.e. a certain element is dismantled and samples of fuel elements are taken which, after radiochemical manipulation, are finally analyzed by mass spectrometric methods.

A review of possible nondestructive methods indicates the only one involving low cost is the measure of the integrated total neutron flux by means of activation of special wires (e.g. cobalt) placed near the fuel. It is recognized, however, that this approach too has difficulties, especially in that there seems to be no simple way to take into account the influence of the flux spectrum which, in a large reactor varies over the distribution of the fuel elements and besides the results are dependent on the variation of the power of the reactor with time.

To avoid, at least in part, this difficulty, it has been proposed to replace the wire with a film of uranium to be attached on the surface of a fuel element and to measure, after the fuel element has been discharged, the quantities of uranium-235 burned-out and plutonium produced in the film itself by means of their alpha spectra.

Of course, the above values, once determined, should be correlated with the quantities of the same isotope inside the fuel elements and this requires the establishment of a correlation theory.

Purpose of the present work is to investigate the feasibility of the method proposed, i.e. to find out under which conditions and with which accuracy it is possible to determine the shape of the alpha spectra, both in unirradiated and irradiated uranium films.

Procedure

The method investigated consisted of the irradiation of thin uranium films contained in suitable boxes attached at the surface of the fuel elements and the subsequent counting of the spectrum of the alpha emitters when the elements are discharged. The ratios of the areas of the uranium-235 and plutonium lines with the area of the uranium-238 line should determine (taking into account the disintegration constants also) the concentration of uranium-235 and plutonium. The values can afterwards be correlated with the amounts of these isotopes in the fuel element itself by means of theoretical calculations.

Two major difficulties have been overcome during this work. First, because of the recent development of semiconductor detectors for alpha particles, resolution values in the range of 0.8-0.4% were attained for the alpha spectra of uranium. Second, it was found that alpha spectra measurements on irradiated 20% enriched uranium were influenced by the high beta background due to the activation of the uranium film. This background caused such an enlargement of the alpha lines that there was no possibility of distinguishing the peaks due to the various isotopes. To eliminate this background, methods of reprocessing the uranium film have been developed with good results.

Based upon the developmental work, a practical application of the method would consist of the following steps:

1. Choose for film preparation uranium of such enrichment, based on foreseen burn-up, that the post-irradiation enrichment will be approximately 14%.

2. By evaporation under vacuum, deposit a layer of the uranium approximately 200 Å thick on high purity aluminum discs of 20 mm diameter and 1 mm thickness.

3. Determine the U-235/U-238 ratio using semi-conductor detectors.

4. Place a disc in a suitable container which is then sealed and leak tested.

5. Attach sealed container to fuel element.

6. After irradiated fuel element is removed from reactor, open container and remove disc.

7. Separate the uranium and plutonium by dissolving in nitric acid and extracting with TBP.

8. Count the samples using semi-conductor detectors and determine the ratio of the interesities of U-235 and Pu line with the U-238 line.

9. Correlate, by theoretical calculation, the U-235 and Pu present in the specimen (disc) with the quantities of the same isotopes within the fuel element.

Conclusions

Considerable work has been carried out in studies and researches with the aim of assessing a method to measure with the required accuracy the alpha spectra both in unirradiated and irradiated uranium.

At this point we can state that the two principal difficulties (the high resolution required and the reduction to tolerable levels of the beta background of the irradiated uranium) have been overcome and a possible sequence of operations for the practical application of the method has been established.

"We can state that the work performed at our Laboratories has lead to the development of equipment and methods suitable for the analysis of uranium-235 and plutonium by means of alpha spectrometry. The application of these methods to fuel elements rests on the possibility of obtaining a correlation between the quantities of those isotopes in the specimens attached externally on the fuel elements and the quantities of the same isotopes in the fuel elements. We think that this correlation can always be obtained by calculations knowing the detailed characteristics of each particular reactor".

We would like to recommend that a realistic experiment be done in which a fuel element is irradiated in conjunction with uranium films so as to make an experimental correlation between the results from the films and the fuel element itself. This experiment would be the natural continuation of the present research.

7. Research Contract No. 92 $ 30,000

Title

A Feasibility Study of Nondestructive Assay of Plutonium-239 in Irradiated Fuel Rods Using Slowing Down Time Spectrometer

Institution: Atomic Energy Establishment
Trombay, India

Principal Scientific Investigators: R. Ramanna, P. K. Iyengar

Period of Contract: 12/1/60 to 9/1/65

Background & Principle

It is known that in case of heavy moderators and for sufficiently high neutron energies, the neutron distribution is given by a Gaussian form, with a constant dispersion. However, when the neutron energy approaches the energies associated with thermal motion and chemical binding, the dispersion is no longer constant and is a function of energy.

Procedure

Using a 400 KV neutron source and 200 cm^3 lead assembly, a lead spectrometer has been constructed. The energy calibration and the slowing down properties both above and below 1 ev were investigated using capture gamma rays from resonance absorbers such as Ag, Au, Ho, Tb, In, Cd etc.

U-235 and Pu-239 have similar fission cross section between .1 ev to 1 ev except at .3 ev where Pu-239 has a resonance whose value is about 20 times that of U-235. Two samples of plutonium of about 150 mg/cm^2 and 45 mg/cm^2 were used in the feasibility studies. It was seen that even with a thick sample and isotropic neutron distribution, it was possible to resolve the fission peak at .3 ev. By comparing the fission rate of Pu to that of U-235 present in the natural uranium, it was possible to estimate the number of plutonium atoms within 20%. The present accuracy in estimation of Pu can be improved, using better counting statistics.

In this method of analysis, the thermal motion influences the slowing down process at the plutonium resonance energy of .3 ev. The measurements, undertaken to study the slowing down process both as regards mean slowing down time and dispersion, indicate that the energy range of a lead slowing down spectrometer can be extended to energies of the order of .2 ev provided the effects of thermal motion and chemical binding are taken into account.

Because of the lack of an intense neutron source, the method could not be applied to the case of an irradiated sample. However, from the data obtained with the plutonium sources, a method of analysis that could be followed in the case of irradiated samples is outlined.

Conclusions

A new approach to the problem of nondestructive assay of fissile material has been investigated and it has been shown that by using a slowing down time lead spectrometer and making use of the difference in fission cross section of U-235 and Pu-239 in the 0.1 ev to 1 ev region, it is possible to estimate their concentration in case of irradiated samples.

A method of estimating U-235 burn-up and Pu-239 production in case of irradiated sample is outlined. This is based on studying the fission yield at two energies besides the resonance energy.

8. Research Contract No. 163 $ 29,000

Title

Nondestructive Analysis of Irradiated Fuel Elements by Establishing a Reliable Correlation Between One or More Gamma Lines From the Fission Products and the Burn-up of Irradiated Fuel Elements or Samples

Institution: Chalmers University of Technology
Gothenburg, Sweden

Principal Scientific Investigators: N. Ryde, J. Rohlin

Period of Contract: 12/10/62 to 4/25/65

Background & Principle

The research project undertaken under this contract concerns a nondestructive analysis of irradiated reactor fuel elements by establishing a reliable correlation between one or more gamma lines from the fission products and the burn-up of irradiated fuel elements or samples.

The methods used for nondestructive burn-up analysis of reactor fuel elements have been intended for technical as well as for control purposes. For both purposes it is desirable to have a reliable nondestructive technique that is applicable to full scale fuel elements which have been subjected to high irradiation doses, i.e. particularly elements from power reactors. For control purposes it is also highly desirable to have a method that can be applied at an arbitrary time of radioactive cooling of the fuel elements. In this connection only long irradiation times of the order of one or several years are of current interest unless very special studies are made. By comparison of the different methods it must be kept in mind that the fact that strongly irradiated fuel elements from power reactors are to be investigated means that even an instrument of very low total efficiency can be used. It is reasonable to believe that the intensity will be sufficient to make it possible to study gamma lines from several fission products and this has been established in this investigation. Instead the highest possible resolving power must be used owing to the complex nature of the radiation emitted from the fission products. If one or several gamma lines from the fission products are used for the burn-up calculations, a resolution of about one percent is necessary for energies below 1 MeV.

Procedure

A review was made of the methods used up to now for the nondestructive analysis in question. The characteristics of the methods are as follows:

Method	Notes
1. NaI spectrometer	Multichannel technique Easy to use Difficult background problems
2. Solid state Compton spectrometer	Multichannel technique Compact spectrometer unit Complicated electronics Difficult background problems
3. Magnetic Compton spectrometer	Point-by-point method Heavy machinery necessary
4. Bent crystal spectrometer	Point-by-point method High resolution Easy to use Moderate machinery
5. 3-crystal scintillation pair spectrometer	Multichannel technique Compact spectrometer unit Complicated electronics Difficult background problems

Beside the methods listed above, there are other possibilities which either have not yet been tried or have proved to be less suitable for nondestructive burn-up analysis of fuel elements.

They are:

1. Germanium (Li) Spectrometers
2. Flat Crystal Diffraction Spectrometers
3. External Gamma-electron Conversion Methods (Using a Double Focusing Magnetic Beta-ray Spectrometer)

A brief discussion of these methods is presented.

It is well known that the U-235 content and the burn-up of fuel in a fuel element can be determined by measuring the intensity of one or several gamma lines from fission products of known fission yield. Owing to the extremely complicated spectrum, a high resolution instrument is needed for this analysis. Recognizing the necessity to resolve gamma lines in the energy region of interest, it was decided to build a line source diffraction spectrometer. Such an instrument should also have a particular merit in permitting studies of very narrow parts of a fuel rod or plate.

The design details are presented for a large crystal diffraction spectrometer having a crystal radius of 4 meters.

Conclusion

By measurements on a small sample of reactor fuel subjected to a burn-up of 270 MWd/t UO_2 it has been clearly demonstrated that it is possible to see gamma lines from fission products with a bent crystal spectrometer. Gamma lines from the decay Zr-95 $\longrightarrow$ Nb-95 $\longrightarrow$ Mo-95 were found. Due to the very short irradiation time of 8 days no attempt was made to find the 661 keV line in the 28 years decay Cs-137 $\longrightarrow$ Ba-137. Neither was any attempt made to estimate the burn-up from the gamma lines measured, because these originate from decays of short half-lives. From the experimental results it could be calculated that burn-up measurements at several gamma lines are possible to perform when a sample of a long-time irradiated full scale element is investigated.

9. Research Contract No. 180 $ 48,800

Title

Nondestructive Analysis of Irradiated Fuel Elements by
Means of Compton Recoil Spectrometry

Institution: Österreichische Studiengesellschaft für
Atomenergie
Vienna, Austria

Principal Scientific Investigator: P. Weinzierl

Period of Contract: 12/17/62 to 12/16/64

Background & Principle

A method was to be developed which allows of the nondestructive determination of burnup of irradiated fuel elements. Gamma spectroscopy is well suited to this, especially if a measurement of ^{137}Cs is obtained. Due to the 30 y half-life of this isotope an effective integration over the irradiation history of the fuel is achieved for the commonly found burning and cooling times. Due to other intensive gamma lines in the neighbouring fission product spectrum (^{95}Zr-^{95}Nb), the measurement of ^{137}Cs requires a high resolution gamma spectrometer, otherwise excessively long cooling times of the order of one year would be necessary.

Procedure

An energy resolution of about 2% for the 662 keV line of ^{137}Cs is required to solve the problem stated above. When the contract was started, only one gamma spectrometer existed which met this requirement, a large diffraction spectrometer made to very stringent mechanical specifications and of high cost.

The use of a Compton spectrometer using a new large type of semi-conductor detector as scattering crystal was suggested by the contractor. The main technological problem was to obtain a semi-conductor detector with a depletion layer of a few millimeters thickness and a homogeneous charge collection characteristic. The first detector produced on special request met the latter requirement only in an unsatisfactory way. But the feasibility of a semi-conductor Compton spectrometer with the required resolution could be demonstrated. During the period of this contract three continuously improved experimental set-ups were developed. The last one had already the form of a very compact, transportable and completely-transistorized system. It contained a spectrometer head with heavy metal collimator, a lithium-drifted silicon detector (3 mm depletion layer) cooled in vacuum to liquid nitrogen temperature and a NaI-crystal with a through-hole for the detection of the backscattered Compton-quanta. A special preamplifier and amplifier system was used for the semi-conductor detector channel. A commercial amplifier and coincidence system with double delay-line shaping and zero crossing timing operation was used and the coincidence spectrum was stored in a multi-channel analyzer.

A scanning machine for MTR fuel elements was installed in the ASTRA reactor pool together with a collimator through the wall into an adjacent chamber. Calibrations and routine measurements on MTR fuel elements and other fuel probes were carried out.

Conclusions

A resolution between 12 and 15 keV (full width at half maximum) was obtained with the last set-up. This allows a ^{137}Cs determination in a fuel element after two weeks of cooling (or longer).

A quantitative check of this burnup measurement method was carried out as follows: the irradiation of a fuel element was first determined by the activation of 15 cobalt wires distributed within the element during irradiation. It yielded a burnup of 37.6 mg ± 7% ^{235}U. The scanning of the element by means of the Compton spectrometer resulted in a burnup of 36.1 mg ^{235}U. Although this agreement is better than the statistical and other errors, an overall accuracy of 10% of this method seems to be a conservative estimate.

The use of a semi-conductor detector Compton spectrometer measuring the ^{137}Cs line allows a nondestructive burnup determination with an accuracy of about 10%.

10. A Gamma-Ray Scanner for Safeguards Use (10/30/59) WCAP-6030

C. C. Webster

A gamma-ray scanner can be used to identify *irradiated* power reactor fuel assemblies by studying their gamma activity, under thirty or more feet of water, with a shielded scintillation detector. Several AEC - supported laboratories have investigated and used similar methods to map the thermal neutron flux in a reactor and to predict the burn-up of fuel elements. The experimental results described show the principles of such a measurement.

A scanner has been designed for use in a Nuclear Materials Control System (NMCS). The device is designed to accomodate fuel configurations ranging in size from a Yankee Power Reactor assembly down to an MTR element, by interchanging an adapter. Basically, the device consists of an arrangement for holding and rotating the fuel assembly, and a shielded movable scintillation detector. The design includes a suitable motion and position control system, and the nuclear instrumentation required to make the specified measurements. The estimated cost of the device including controls and instrumentation is about $20,000.

The scanner was designed for use in a spent fuel storage pit. The materials of construction were chosen to minimize contamination of the water in the storage basin. The nuclear instrumentation, the electrical control unit, and the drive motors would be located at the edge of the pit. The electrical control unit regulates all the motions of the scanner and is fail-safe.

A mock-up of the electrical control system was fabricated and tested. The nuclear instrumentation system also was assembled and tested. The nuclear instruments included a specially designed transistorized preamplifier which was built and evaluated.

The scanner will fulfill the NMCS requirements for identification of *spent* fuel elements before and after shipment. The device can also be used to assist in allocating the calculated fissile material content of spent reactor cores to individual fuel assemblies. A feasibility study on the use of the device to determine the absolute fissile material content of spent power reactor fuel assemblies indicates little promise of success.

11. The Fuel Assay Scanner (2/1/60) WCAP-6039

R. V. Babcock, S. L. Ruby

For the purposes of the Nuclear Materials Control System (NMCS), a device was required which would perform a non-destructive assay of the materials in a completed reactor fuel assembly. As a practical example of such a machine, a system was developed to assay the U-235 and poison (boron) content of high-enrichment plate-type fuel elements by comparison with a standard element.

The NMCS prototype device is capable of assaying the U-235 content of ETR and MTR fuel elements with an absolute accuracy of 0.3% (95% confidence level) by the measurement of the natural gamma activity. ETR elements containing boron may in addition, be assayed for boron content with a relative accuracy of 4% by a combination of the above gamma-ray technique, thermal neutron transmission and a volume determination. The individual measurements require less than 15 minutes.

An epi-thermal neutron transmission measurement was also developed as a means of distinguishing between boron and U-235, but was found to have inadequate counting statistics for assay applications.

Consideration has been given to use of the above methods for various assay problems. Preliminary recommendations of combinations of methods to be applied to specific assay problems have been made.

The instrument was calibrated absolutely by the chemical analysis of fuel plates which had been assayed by the gamma-ray technique. The resultant calibration curve was employed to determine the U-235 content of other plates which were in turn assembled to form a series of permanent standard fuel assemblies.

The fuel element assay device has been adequately engineered so that normal changes in operating parameters cause errors which are small compared with those quoted. The machine is deemed ready for field testing.

12. Field Testing of the Fuel Assay Scanner (6/22/60) WCAP-6042

F. J. Arsenault

A Fuel Assay Scanner was designed and constructed by Westinghouse personnel for the non-destructive assay of MTR and ETR type fuel elements. The instrument was designed to assay uranium-235 by measuring the natural gamma-ray emission, and to measure uranium-235 plus poison content by a thermal neutron transmission technique. A combination of the two measurements allows calculation of the poison content. The Scanner was shipped to the Materials Testing Reactor at the National Reactor Testing Station for evaluation under typical field conditions. Eighty MTR and eight ETR fuel elements were scanned, and a preliminary calibration of the boron assay was accomplished. Uranium-235 content may currently be assayed with the Scanner to ± 0.54%; the boron content may be measured to ± 0.1 grams. Measurements on the aluminum content of the elements are expected to improve the accuracy to ± 0.21% for the uranium-235, and ± 0.06 grams for the boron. The Scanner has been transferred to the Phillips Petroleum Company, and is currently in operational use at the MTR-ETR site.

13. A Method for the Nondestructive Analysis of Power Reactor Fuel Elements (7/22/60) WCAP-6044

B. Jennings

A technique for performing a non-destructive assay for U-235 on large low enrichment power reactor fuel assemblies has been developed for use in a Nuclear Materials Control System (NMCS).

The primary problem in the development of a suitable assay procedure is to obtain adequate assay sensitivity to fuel in the interior of the element under test. The method chosen to achieve this penetration is to bombard a moderated fuel assembly with fast neutrons. The fast neutrons penetrate into the fuel assembly, are moderated by the water, and produced thermal and resonance fission in U-235. In order to minimize the penetration required, the fuel assembly is rotated about its major axis so that the fast neutron source irradiates a short length of the assembly at each assay. A measure of the number of fission events, and hence U-235 content, is obtained by counting the number of delayed neutrons released in the fission process.

The above technique has been verified experimentally using 2.6 Mev neutrons from a Cockcroft-Walton generator and measuring the delayed neutrons produced in a Yankee-type fuel assembly. The results show that the number of delayed neutrons for constant fast neutron activation is directly proportional to the uranium in the assembly, except for the inner 10% where the sensitivity decreases.

14. Research Contract No. AT(11-1)-1321 $ 20,000

Title

Nondestructive Assay of Nuclear Fuels

Institution: Southwest Research Institute

Principal Scientific Investigator: W. J. McGonnagle

Period of Contract: 8/15/63 to 5/26/64

Background & Principle

The determination of the uranium and plutonium content of unirradiated fuels is of interest to the reactor physicist and to those in charge of accountability of uranium and plutonium materials. The reactor physicist is interested in the total amount of fissionable material in the fuel elements and the homogeneity of the loading. The accountability offices are interested only in the total amount of fissionable material in a particular fuel element or sub-assembly and are not concerned with the distribution.

The amount of burn-up in a reactor fuel element is also of consideration to the reactor physicist and people responsible for accountability. Reactor physicists want to know the amount of power produced as a function of burn-up. The accountability personnel are interested in determining how much plutonium has been produced.

Objectives

1. To determine the feasibility of using a modified Compton magnetic spectrometer for analyzing the uranium and plutonium content of cold and irradiated nuclear fuels.

2. To evaluate the use of solid state detectors in gamma ray spectrometry for both cold and irradiated nuclear fuels.

3. To devise methods and techniques for determining the special material content of scrap.

Conclusions

This study, supported by the Division of Nuclear Materials Management, resulted in a report containing the following conclusions and recommendations:

1. The Compton magnetic spectrometer is too inefficient to be used in fuel assay on a production line basis.

2. Simple experiments should be performed to substantiate that the induced fission technique is a practical technique of fuel assay.

3. The induced fission technique and the X-ray transmission technique can be used for analyzing the fissionable material content and enrichment of reactor fuel plates.

4. The neutron transmission technique should be experimentally investigated for use in determination of the fissionable material content of scrap material.

5. The use of solid state detectors should be tried on cold fuel as well as hot fuel plates. The effect of radiation damage on such detectors must be determined experimentally.

6. Solid state gamma detectors operating at 77°K should be evaluated for use in fuel assay.

7. The use of solid state detectors, with their higher resolution, should be used to scan the spectrum of uranium in order to get a better idea of the energies present in the spectrum and the origin of these energies.

15. Research Contract No. $ 57,000

Title

Nondestructive Analysis of Burnup in Irradiated Fuel Elements

Institution: Arthur D. Little, Inc.

Principal Scientific Investigators: R. H. Johnston, E. A. Vrablik, N. C. Rasmussen

Period of Contract: 1963 - 1964

Background & Principle

In connection with efforts to determine the best methods for assaying nuclear fuel, it was necessary to determine the feasibility of measuring the uranium to plutonium ratio nondestructively. Accordingly, the Division of Reactor Development awarded a contract which resulted in the preparation of a report, "Nondestructive Analysis of Burnup in Irradiated Fuel Elements," dated April 17, 1964.

Objective

Determine the feasibility of measuring the uranium to plutonium ratio in air.

Procedure

At the outset of this problem it was envisaged that all testing of feasibility would be carried out in air. The initial investigation of the feasibility of this approach was carried out at the Massachusetts Institute of Technology using a 2-meter-bent-quartz-crystal spectrometer of the DuMond type and examining a fuel rod from the MIT reactor. Identification of the lines was initially done by means of a photographic film, though it was recognized that this would prove too slow for any practical equipment.

An instrument was designed to assay fuel and was shown to have the necessary resolution for measuring the uranium to plutonium ratio in laboratory tests. The instrument failed to resolve the fluorescence lines when used under water in examining spent fuel assemblies of Core I at Yankee Atomic Electric Company. This failure is attributed to a defect in shielding design.

Results

The initial objective of this program, the feasibility experiment conducted in air as originally intended, has been realized. The modified objective of carrying out the tests on Core I at Yankee Atomic Electric Company reveal deficiencies in the instrument for operational use.

16. Research Contract No. W-7405-ENG.36

$ 748,000
to 9/30/68

Title

Pulsed Neutron Research for Nuclear Safeguards

Institution: Los Alamos Scientific Laboratory

Principal Scientific Investigators: G. R. Keepin and Group N-6 Staff

Period of Contract: 1/30/67 to (continuing)

Background & Principle

The implementation of an effective nuclear safeguards and materials management system requires direct physical methods of detecting, identifying and quantitatively analyzing fissionable materials in various practical configurations containing both fissionable and nonfissionable materials. To be most effective and useful, such assay methods should be nondestructive, rapid, accurate, and capable of being carried out under a wide range of both laboratory and field conditions; e.g., in situ or on-line assay in materials processing plants, or in mobile isotopic assay stations for in-the-field use. The difficulties of representative sampling in (destructive) chemical assay of heterogeneous and complex systems are largely obviated in new nondestructive assay methods, largely because of their characteristic high penetrability through bulk materials.

Nondestructive assay methods can be divided into two main categories: (1) passive assay, and (2) active interrogation. Passive assay methods involve observation of both neutrons (from spontaneous fission or (α,n) reactions) and gamma rays (emitted following α decay) which are uniquely characteristic of individual fission species. The naturally occurring gamma lines having sufficient intensity for passive assay applications are typically a few hundred kilovolts or less in energy and hence have limited penetrability through dense materials. For many practical assay problems this lack of penetrability severely limits the usefulness of passive methods, and one must employ active interrogation techniques. From an inspection and surveillance standpoint, it may also be noted that active interrogation techniques are inherently more difficult to subvert or circumvent than are the simpler passive techniques.

Active interrogation involves the use of an external source of neutrons or photons to induce fissions in the material under investigation. Neutron sources were chosen for active interrogation in the LASL safeguards program because of (1) higher effective penetrability of fast neutrons in nuclear materials generally; (2) sharp, well-defined neutron fission thresholds which provide incisive isotopic discrimination; (3) readily- available, simple, inexpensive, compact neutron sources (e.g., D-D, D-T neutron generators, Cf-252, etc.) of the required intensity and reliability for practical assay applications; and (4) the expectation of smaller radiation dose (RAD) delivered to the unknown sample for a given assay capability.

In active interrogation methods, quantitative assay is based on detailed observations of the delayed and prompt neutrons and gamma rays from fission. The delayed regime has the advantages of complete time-separation from the interrogating pulse, and permits the use of simple, inexpensive counting circuitry and associated data reduction equipment. Emphasis at LASL is placed on the development of very high efficiency detectors so as to minimize the interrogating-neutron-source strength with its associated shielding requirements, personnel hazards, etc.

The characteristic differences in yields and kinetic response of the delayed neutrons from the various fission species provide a unique method for analysis of individual isotopes in unknown mixtures of fissionable and nonfissionable materials. The experimental techniques involved in delayed neutron assay are rapid, nondestructive, and relatively simple and inexpensive. For relative isotopic assay, no absolute calibrations (of source, detectors, fission rates, etc.) are required, and all measurements are made in a time domain which is completely free of perturbations due to prompt neutron higher modes, diffusion and thermalization effects.

In addition to delayed neutrons, delayed fission gamma rays and prompt fission neutrons and gamma rays may also be used to provide characteristic signatures of individual fission species. New nondestructive assay techniques developed on the basis of these signatures can be intercompared with the conventional chemical assay methods in use for many years in the various pilot-plant operations at LASL which represent essentially every stage of industrial fabrication, recovery, and processing of nuclear materials. Full exploitation of the various passive and nondestructive assay methods under development at LASL promises a wide range of applications to safeguards and nuclear materials accountability problems throughout the nuclear industry.

Objective

The purpose of this program is to explore and develop promising new approaches to the nondestructive detection, identification, and analysis of fissionable materials, as well as to improve on assay methods used in the past. The detailed kinetics, energy and yield characteristics of delayed and prompt fission neutrons and gamma rays provide the fundamental signatures for various fissionable materials. These basic data are being applied in the development of complete systems for nondestructive isotopic assay, and the early application of such systems to practical safeguards and nuclear materials management and accountability problems in the expanding nuclear industry.

Procedure

The program will consist of the simultaneous development of both passive and active interrogation methods for the nondestructive isotopic assay of fissionable materials. The early implementation of practical assay systems requires development of high efficiency neutron and gamma detectors and the adaptation of compact, commercially-available neutron sources to insitu or mobile "in-the-field" assay systems.

The key steps foreseen in the LASL program are:

1. Measurements of fundamental fission signatures whenever existing data are inadequate for assay applications. The required basic measurements include: absolute delayed neutron yield versus energy of the neutron inducing fission, time-dependent energy spectra of delayed neutrons, group abundances and decay constants of delayed neutrons for 14 MeV neutron induced fission, delayed gamma-ray lines characteristic of individual fission species (using high resolution Ge(Li) gamma detectors) which have suitable intensity and energy for assay applications.

2. Basic computational and experimental studies of neutron moderator configurations for "spectrum tailoring" of available high-energy neutron sources (e.g., D-D, D-T, Cf-252). The use of spectrum tailoring shifts a large portion of the source neutrons below the thresholds of the fertile isotopes (U-238, Th-232), thus providing enormously increased isotope discrimination factors.

3. Development of practical assay methods through experimental measurements and parallel theoretical calculations of delayed neutron and gamma-ray response of various practical configurations (e.g., fuel pellets, fuel assemblies, and scrap) to pulsed neutron interrogation.

4. Development and adaptation of experimental equipment (e.g., detectors, accelerators, sample handling devices) required for safeguards R&D. The Van de Graaff accelerator as a variable-energy source of monoenergetic neutrons will be fully explored for nondestructive assay applications.

5. The new nondestructive assay methods and results for typical scrap, fuels, etc. produced at LASL and other nuclear facilities will be directly intercompared with destructive chemical assay to be carried out at LASL and elsewhere.

6. Adaptation and integration of proven new assay methods into a mobile assay system (roadable van) suitable for practical field use by the AEC. Undoubtedly the new technical methods being developed for safeguards inspection and control will find many practical commercial applications in the area of materials management, e.g. quality control of process lines and fabricated product (new fuel elements), on-line scrap monitoring, rapid nondestructive assay of burnup in spent fuel elements.

Status as of 3/1/68

(From BNL report of visit to LASL on February 21 and 22, 1968, by H. Kouts, C. Sastre, R. Chrien, and W. Higinbotham)

At the present time Los Alamos Scientific Lab. has an active program of research, both theoretical and experimental. This program leads to assay techniques for both hot and cold fuel elements and waste drums and naturally will produce the basic information and theoretical understanding required to fundament such techniques.

As part of the technique development, a device or possible various devices will be constructed as prototypes. One device could be finished in the present calendar year, but its characteristics are not yet well defined.

Laboratory testing has started on MTR type fuel elements and they will extend the testing to various other designs. They have started collecting information on waste drums from LASL to help on future tests.

Considerable attention has been given to the availability, cost and transportability of devices based on accelerators and radioactive sources.

Simple calculations of sensitivity for detection of small amounts of uranium had encouraging results.

As a complement to the development of the techniques, they are investigating the properties of the delayed neutrons from high energy fission. This work is valuable not only for safeguards applications but is useful for the industry in general.

Status as of 4/1/68

Kinetic Response Techniques for Nondestructive Assay of Fissionable Materials

During the first quarter of 1968 research on kinetic response techniques progressed from experimental studies on small, rather idealized, samples to more practical, complex and heterogeneous geometries as for example MTR fuel elements and fissionable materials in scrap barrels.

Nondestructive Assay of Reactor Fuel Elements

In order to investigate the accuracy and practicality of delayed neutron techniques for measuring absolute amounts of fissionable material in reactor fuel elements, a series of measurements have been performed on an MTR-type fuel element mock-up. The resulting measured isotopic abundance agreed with the actual isotopic abundance to within 1% (0.4% in the case of 14 MeV neutron interrogation and 0.7% in the case of moderated-spectrum neutron interrogation).

Fissile Material Assay in Scrap Barrels

An important practical problem in special nuclear material accountability is the determination of the amount of fissile material in common scrap containers. Delayed neutron kinetics and yield measurements offer a promising method for quantitative scrap assay. Basically, the method consists of irradiating the scrap container with a modulated neutron source and then measuring the delayed neutrons resulting from the induced (n,f) reactions in the fissile material. Preliminary measurements have been performed to determine the sensitivity for detecting a small amount of fissile material in a standard 55-gallon steel barrel. It is planned in future measurements to include representative absorbing, moderating, and scattering materials interspersed with fissile material in scrap barrels. These effects are now being calculated in detail using neutron transport computer codes.

Absolute Delayed Neutron Yield Measurements

During the past quarter several improvements and modifications have been made to the absolute delayed neutron yield measuring techniques described in earlier progress reports. In addition, yield-vs-energy measurements have been extended to the Pu-239 and U-233 isotopes. The major modifications include (1) introduction of a new high-efficiency long counter employing five He_3 detectors rather than one BF_3 detector as in the original Hansen-McKibben design; (2) use of fission chamber sandwich to obtain averaged fission rate in the sample; (3) remote swing-away arm for accurate, rapid background determination; and (4) increasing the Cockcroft-Walton accelerating potential to 325 kV to augment D,D neutron output.

Most recent measurements confirm the previously reported decrease in absolute delayed neutron yield in going from 3 MeV to 14.9 MeV neutron induced fission for each of the five major isotopes studied.

Neutron Transport Calculations

Optimum Moderator Configurations

Neutron transport calculations using the DTF-IV transport code have been directed toward maximum neutron leakage and desired spectral characteristics from various moderator assemblies. Calculations were performed in spherical geometry with a 0.5 cm diameter, distributed neutron source at the center of a moderating sphere. Both a 14 MeV source and a fission source were used. Each moderator assembly consisted of concentric spherical shells of various materials with varying radii. The calculated results show in a very striking manner the desired spectrum peaking effect just below the U-238 threshold. (For example, using a 10 cm tungsten sphere around a D,T source, nearly 90% of the 14 MeV source neutrons are shifted below 1 MeV.)

Neutron Response of Simulated Scrap Barrels

An extensive program of neutron transport calculations has been undertaken to guide experimental investigations of nondestructive scrap assay techniques. Present calculations using the one-dimensional DTF-IV transport code are concerned with simulated scrap barrels and the expected delayed neutron response from small amounts of fissile material interspersed in large amounts of scrap. The standard 55-gallon scrap barrel is simulated by a sphere of radius 30 cm--the radius of a standard 55-gallon drum.

These neutron transport calculations point up the very useful general result that barrels containing almost any common scrap material other than hydrogenous material, and weighing up to a few hundred pounds, appear empty, or are essentially "transparent" to 14 MeV neutrons. Even the hydrogenous materials, when present in low densities (e.g., in "light" scrap barrels containing paper trash, rags, filters, etc.), are essentially transparent to 14 MeV neutrons and should pose no serious problems as regards fast neutron interrogation and assay. It is clear, however, that large amounts (i.e., high densities) of hydrogenous material (if not known to be present, and hence not taken into account) would give greatly enhanced delayed neutron response with a consequent overestimation of the amount of fissile material present in the scrap container. A measurement of the cadmium ratio--the ratio of total neutron counts to those above 0.3 eV--may provide a very convenient indication of hydrogenous scrap present.

Auxiliary Assay Techniques Using Fission Delayed Gamma Rays

Delayed gamma rays from fission may either provide unique isotopic signatures or complement delayed neutron assay methods already developed, depending on the specific gamma-ray characteristics measured. The gross delayed gamma-ray intensity (total photons per fission-sec) over the time range from $\sim$0.1 sec to 100 sec after fission depends on the isotope undergoing fission in much the same way as the delayed neutron emission. The yields of individual early delayed gamma-ray lines are also of interest for safeguards applications; measurements of these gamma lines with high resolution Ge(Li) detectors are presently underway at LASL. In addition, isotopic assay techniques based on delayed gamma-ray characteristics are currently under active development.

Detector and Instrumentation Development

Two versatile, high-efficiency neutron detectors of the slab design have been constructed and are presently being used for applications requiring large area or large-solid-angle neutron detection. These detectors will also be used as coincidence counters to study the feasibility of detecting neutrons from Pu-240 spontaneous fission in a background of (α,n) neutrons.

Fabrication of the high-efficiency 4π neutron detector has been completed except for assembly of the shield and wiring of the high-voltage junction boxes.

A general purpose charge sensitive preamplifier using field-effect transistors has been developed for use with proportional, solid state, and scintillation detectors.

Dense Plasma Focus Source

The Dense Plasma Focus (DPF) source, which was completed during the previous reporting period, is now undergoing an intensive program of testing, debugging, and modification, with the intended goal of increasing total neutron burst yield, reproducibility of yield, reliability, and ease of operation.

In the period from 1 January to 9 February, the DPF was fired 247 times. Of these, 81 shots gave measurable yields varying from 5×10^6 to 4.8×10^9 neutrons in bursts of 50-100 nanoseconds width.

Accelerator I Neutron Source

Accelerator I, a compact, mobile neutron source was delivered to LASL on January 15, 1968. The source is currently undergoing stringent performance tests and evaluation for safeguards assay applications.

Status as of 7/1/68

Neutron Interrogation Techniques for Nondestructive Assay of Fissionable Materials

The delayed neutron kinetic response measurements reported in previous progress reports have been extended to include mixtures of Pu-239 and U-238. Overall measured isotope discrimination factors of 5 or greater between U-238 and Pu-239 have been demonstrated. Further delayed neutron yield and kinetic response measurements on prototype FBR fuels are planned during FY 69 and subsequently.

Nondestructive Assay of Reactor Fuel Elements Using a "Fast-Tailored" Neutron Beam

In the practical assay of many types of reactor fuel elements it is desirable to interrogate with neutrons that have energies high enough to be very penetrating, but which lie below the thresholds (several MeV) for (n,p), (n,2n), and (n,n'f) reactions. This desired result can be readily achieved by "spectrum tailoring" of 14 MeV (D,T) neutrons, using an 8" -thick Pb slab to reduce the average neutron energy from 14 MeV to a few MeV.

Delayed neutron response measurements have been performed on an MTR-type fuel element mockup to determine the absolute amounts of fissionable material in elements with various loadings. The mass of fissionable material determined from the delayed neutron response agreed closely (average deviation = 0.51%) with the actual mass. A particularly significant result of this work is the fact that absolute total amounts of fissile material can be measured to within 1% accuracy when appropriate calibration standards are used. Thus the delayed neutron method of nondestructive assay seems very well suited to the determination of absolute amounts of fissionable materials in various types of reactor fuel elements.

MSRE Salt Assay be Delayed Neutron Kinetic Response Methods

At the request of Oak Ridge, a series of experiments is in progress at LASL to develop neutron kinetic response methods for nondestructive, quantitative assay of U-233 and Th fuel-salt mixtures to be used in the Oak Ridge molten salt reactor experiment (MSRE reactor).

A typical MSRE salt contains Be, F, and Li in addition to the fissionable nuclides, U-233 and Th-232.

Present measurements give a U-233/Th delayed neutron response ratio between 24 and 170 (corresponding to tailored spectrum assemblies with and without Cd). Considerable further improvement in U-233/Th fission ratios is expected using more nearly optimum spectrum tailoring configurations.

Neutron Interrogation and Delayed Neutron Measurements Applied to Scrap Assay

As has been pointed out in previous progress reports, the presence of hydrogenous material in fissile scrap configurations yields greatly enhanced delayed neutron response. One method of reducing this response is to normalize observed delayed neutron counts to fission counts in a fission chamber monitor containing the fissile species of interest. This has proved a most effective means of compensating for the presence of hydrogenous materials in practical scrap configurations. Both calculations and experiments are being extended to more complex arrangements of sources and detectors, as well as different slab thicknesses, and to neutron source spectra other than 14 MeV. These studies are directed toward the development of reliable, accurate neutron interrogation techniques for quantitative assay of a wide range of practical scrap configurations.

Neutron Response of Simulated Scrap Barrels to Fast-Tailored-Spectrum Interrogation

The neutron transport study of simulated scrap barrels, begun early in 1968, has been extended to investigate the delayed neutron response to a fast tailored source produced by a 10 cm thick tungsten moderator around a 14 MeV source.

As in the case of the 14 MeV source, barrels containing almost any common scrap material other than hydrogenous material and weighing up to a few hundred pounds are essentially transparent to this fast-tailored-spectrum neutron source.

Absolute Delayed Neutron Yields

The program of absolute delayed neutron yield measurements from 3.1 and 14.9 MeV neutron-induced fission of the major fission species was completed during the second quarter of 1968. These measurements show clearly that the delayed neutron yield of every isotope studied decreases significantly in going from 3.1 MeV to 14.9 MeV neutron-induced fission--a result which is expected from the known behavior of fission mass and charge distributions as a function of incident neutron energy in neutron-induced fission. As noted previously, these results stand in direct contrast to previous measurements by other workers both in the U.S.A. and the U.S.S.R.

A summary of the LASL delayed neutron absolute yield measurements was reported at the June, 1968, meeting of the American Nuclear Society in Toronto.

Accurate absolute delayed neutron yields as a function of incident neutron energy are essential to the practical application of delayed neutron response techniques to nondestructive isotopic assay.

Self-Indication Techniques for DIA Applications

The possible use of the resonance self-indication technique for nondestructive assay of fissile materials has been investigated. This technique utilizes the characteristic resonance structure in the neutron fission cross sections of the fissile isotopes. A collimated neutron beam from the LASL Water Boiler Reactor was passed through a Gd foil (4.5 mil) or a Cd foil (30 mil) to remove the thermal neutrons, then passed through the fissile sample under investigation, and finally through a sandwich of three parallel-plate ionization chambers containing thin evaporated deposits of Pu-239, U-235, and B-10. Counting rates in all three detectors were recorded as a function of thickness of the various fissile samples in the beam.

These responses represent the neutron transmission weighted by the fission cross sections of the detectors. The results of the measurements show that thicknesses (in the region of $\sim$20 mil) of Pu-239 and U-235 can be determined to within approximately 1% and 2.6%, respectively, by single transmission measurements. Measurements made with a Pu sample (0.010" thick) sandwiched between relatively thick layers of common materials showed (as anticipated) that the ordinary transmission measurement is quite sensitive to the material whereas the changes in the detector ratios are typically only a few percent.

Passive Neutron Counting of a Cold MTR Fuel Element

For reactor elements containing alloys, mixtures, or chemical compounds of fissionable materials and low Z elements, counting of neutrons produced by (α,n) reactions initiated by α-particles from the fissionable materials, as well as neutrons from spontaneous fission, may provide a useful means for monitoring fuel content and material uniformity of cold fuel elements. An experiment was performed to determine if a detectable number of neutrons are emitted from a cold MTR fuel element which contains approximately 340 grams of uranium ($\sim$ 93% U-235, $\sim$ 5-1/2% U-238, $\sim$ 1% U-234, $\sim$ 1/4% U-236) alloyed with aluminum. The results showed that ample neutron counts could be obtained within a few minutes for MTR elements, if a high efficiency ($\sim$ 70%) 4π detector is used.

Coincidence Counting of Neutrons from the Spontaneous Fission of Pu-240

To measure the sensitivity of the N-6 slab detectors for coincidence counting of neutrons from the spontaneous fission of Pu-240, a 2.356-gram Pu sample (94.15% Pu-239, $\sim$ 5.46% Pu-240) was placed midway between the slab detectors which were positioned face-to-face in close geometry. The coincidence rate was measured using an 80μsec coincidence gate width, and the background (random coincidence rate) was measured by delaying the output of one of the detectors by 500μsec.

The results indicate that the present system can determine Pu-240 concentrations in typical oxide fuels containing U-235, U-238, and Pu-239.

Neutrons from Spent Fuel Elements

The possible use of neutron counting techniques to determine the Pu buildup in spent reactor fuel elements has been investigated in some detail. In order to estimate the neutron production from spontaneous fission and (α,n) reactions due to elements other than Pu-239 and Pu-240 present in an irradiated fuel element, a computer code has been written which calculates the concentration of any desired element from multiple neutron capture and subsequent decay chains as a function of time during and after neutron irradiation.

Calculations have shown that neutron production from spontaneous fission of Cm-242 would be about an order of magnitude greater than from Pu-240 spontaneous fission for freshly irradiated fuel elements. In addition, the neutron production from the spontaneous fission of Cm-244 would be of the same order of magnitude as from Pu-240.

It is concluded from these calculations that the expected neutron yield from a spent fuel element is sufficiently high to obtain an accurate count with high-efficiency neutron detectors. However, since Cm-242, the primary source of the neutrons, has a half-life which is short compared with typical in-pile times, quantitative information on the Pu-239 could be inferred from neutron measurements only if detailed fuel irradiation history is known. Conversely, passive neutron counting of fuel elements which have been irradiated under controlled conditions could be used to verify, nondestructively, the predictions of burnup and transuranic production codes (such as ISOCHECK).

Delayed Gamma Rays From Fission

For active interrogation assay methods utilizing fission delayed gamma rays, the early time domain is expected to be most important because: (a) the largest isotopic differences in delayed gamma-ray characteristics should be found at early times; (b) the gamma-ray energy spectra should be richer in distinct high-energy (penetrating) gamma rays which can be resolved with Ge(Li) detectors. Analysis of gross delayed gamma-ray activity may complement delayed neutron assay techniques because of differences in neutron and gamma-ray penetrabilities in various materials. This may be particularly useful when the environment of the nuclear material is unknown, e.g., high density scrap.

An active neutron interrogation system based on individual early delayed gamma-ray (line) yields should prove useful for safeguards if the fundamental high-resolution gamma-ray spectra show large differences in the yields of high energy gamma rays from U-235(n,F) and Pu-239(n,F). In this case interrogation with subthreshold neutrons could be used to obtain concentrations of Pu-239 and U-235, separately, in the presence of a large quantity of U-238, as occurs, for example, in power reactor fuels.

Detector and Instrumentation Development

Slab Detectors

The N-6 high-efficiency slab detectors have proven to be very useful in various aspects of nuclear safeguards research.

The total energy response of the slab detector is essentially independent of neutron energy from a few keV to greater than 2 MeV. (Typical delayed neutron spectra lie well within this range.) The intrinsic efficiency for detecting a neutron entering the active area (∼20" x 24") of the slab detector is 13%.

Total Absorption Ge(Li) Gamma Ray Spectrometer

A coaxial Ge(Li) "duode" detector with an active volume of ~ 16 cm^3 has been lithium drifted at LASL and is now ready to undergo preliminary testing. Special charge-sensitive preamplifiers for use with this detector have also been designed and built. A detector of this type should provide a valuable laboratory tool for on-line γ-ray counting of fissile materials for isotope identification and practical assay applications.

Dense Plasma Focus Source

The intensive program of testing, debugging, and modification of the N-6 Dense Plasma Focus (DPF) pulsed neutron source has now achieved the desired neutron yield range ($\sim 10^{10}$ D,D neutrons/pulse) with adequate reproducibility and reliability.

Intensive testing has demonstrated that the Dense Plasma Focus is a practical, reliable neutron source, capable of producing neutron pulses of width 50 to 100 nanoseconds, with (D,D) neutron yields the order of 10^{10} —original design objective. It will be recalled that this corresponds to a yield of 10^{12} (D,T) neutrons per pulse for a deuterium-tritium gas mixture replacing the present deuterium charge. Thus the DPF pulsed neutron source is ready for use in appropriate safeguards research problems; the first problem will be a measurement of delayed gamma rays from fission in the interval from 0.10 to a few seconds.

Accelerator I Neutron Source

The N-6 mobile compact neutron source, "Accelerator I," continues to perform with full reliability. Two new quick-change target holders have been designed and fabricated for fast, convenient target replacement in laboratory and field installations.

Both Accelerator I and the Cockcroft-Walton were in almost continuous use during the second quarter of 1968, and both continue to perform very satisfactorily in both the DC and pulsed modes of operation.

17. Research Contract No. AT(04-3)-167

$ 366,000
to 9/30/68

Title

Investigation of Photoinduced Reactions for Nondestructive Nuclear Materials Safeguards Applications

Institution: Gulf General Atomic - Accelerator Physics Department

Principal Scientific Investigator: J. R. Beyster

Period of Contract: 10/1/67 to (continuing)

Background & Principle

The control of fissionable materials is expected to become a very difficult task in the next few years since the use of nuclear power and the production of these materials will increase very rapidly. One limit to the improvement of nuclear materials control is that there are substantial amounts of nuclear materials in the nuclear fuel cycle which are unmeasurable by the conventional sampling and weighing techniques. Some of the unmeasurable material is incorporated into configurations which cannot be subjected to destructive tests, such as fuel elements, or critical assemblies. Other material is in a form unsuitable for sampling, such as very radioactive materials and wastes in fuel reprocessing or very dilute cold wastes from fuel fabrication. So long as there is a significant amount of unmeasurable material, the accounting procedures are subject to deception. To reduce the amount of unmeasurable fissionable material, research and development is being done at Gulf General Atomic on non-destructive measurement of these materials using electron accelerators.

The principal features of electron accelerators that make them useful for nondestructive assay are: (1) a small accelerator can produce a high intensity beam of penetrating gamma rays which produce many characteristic nuclear reactions; (2) the end point energy of the bremsstrahlung spectrum of gamma rays can be varied easily, and the yields of the characteristic reactions change rapidly with end point energy. By measurement of the yields of these characteristic reactions both the amount and isotopic composition of fissionable material can be determined.

The most abundant characteristic products produced in nuclear materials by bremsstrahlung are the prompt neutrons resulting from photofission and gamma-n reactions. The thresholds for these reactions in nuclear materials are, with few exceptions, lower than the thresholds in other materials. Hence, the prompt neutron yields at low energies ($<$ 6.5 MeV) are very promising identifying characteristics. The integrated over time yield of delayed neutrons following photofission is less than the prompt neutron yield by about two orders of magnitude, but this quantity has an important advantage: photons of the energies we are considering produce delayed neutrons only in nuclear materials. Hence, some of the reduction in yield may be compensated by using electron energies higher than those used for prompt neutrons.

The time dependence of the delayed neutrons in the time range of the order of few seconds to one minute following the irradiation of a fissionable material may be used as a signature of that material. The yield in this kind of measurement is considerably lower than the integrated delayed neutron yield since the irradiating source must be turned off for about one minute while the time distribution is measured. Even so, with high fluxes possible from electron accelerators, delayed neutron time distributions may be useful signatures.

Other relatively low yields, similar to the delayed neutron yield in origin, are the yield of delayed gamma rays following photofission and the yield of specific fission product gamma ray lines as measured by a high resolution detector. Assay techniques based on gamma ray lines have the important advantage that they are very specific.

Objective

The objective of this research program is to develop credible assay techniques for quantitative isotopic measurements of special nuclear material in the various physical forms likely to be encountered throughout the nuclear materials fuel cycle.

Procedure

To achieve the objective the following work is to be performed:

1. Review and classify properties of the photofission process insofar as they apply to nuclear materials safeguards problems.

2. Investigate and compare selected active surveillance techniques based on bremsstrahlung beams for use in identification and classification of nuclear materials.

3. Perform basic experimental work on photoinduced reactions (photofission) to provide information essential to the establishment of nuclear materials signatures; and develop computer methods necessary to support these applications.

4. Commence an applications study on the use of active safeguards techniques involving electron accelerator sources.

5. Perform selected laboratory tests using promising safeguards techniques and establish the range of applicability of these techniques.

6. Initiate a study of radiation sources for their suitability for utilization in the test program.

Status as of 10/16/68

Introduction

The experimental program to date has involved the performance of certain key screening experiments to demonstrate the magnitude of the differences observed between the several materials of interest.

A second portion of the program has considered the practical application of the ideas generated in the basic reaction mechanisms studies to practical assay problems. Mock configurations representing large samples containing dispersed fissionable material are being assembled, and a scanning system has been built to perform measurements on these configurations. The bulk sample scanning mechanism is designed principally for assay of scrap barrels but may also be used for other samples, up to 24 in. diameter. It is intended to provide the necessary translation and rotation of the sample to ensure that the assay is equally sensitive to nuclear material located in every volume element of large samples. This device will be tested on the existing electron accelerators at Gulf General Atomic until a prototype accelerator is developed specifically for this application.

Development of Signatures of Nuclear Materials

1. Prompt and Delayed Neutrons

Measurements of the prompt yields have been made on U-238, U-235, and Pu-239 and will eventually be made on Pu-240, U-233, and Th-232. The prompt neutron yields have markedly different energy dependencies for different isotopes. In particular, the prompt neutron yield shapes for U-235 and Pu-239 are quite different. This is important since most other assay techniques do not provide good discrimination between Pu-239 and U-235.

Delayed neutron yields integrated over time have been measured in the same experiment. The delayed neutrons are distinguished from the prompt neutrons by their time of detection. Prompt neutrons are detected within a few hundred microseconds after an irradiating pulse. Since the next pulse occurs in a few milliseconds, the delayed neutrons appear as a uniform distribution in the time interval after about 1 millisec up to the next irradiating pulse.

Since delayed and prompt neutron yields can be measured simultaneously with the same irradiating flux, it is sometimes useful to use the ratio of delayed to prompt neutrons as a relative signature. This type of signature has the advantage of being independent of the absolute magnitude of the flux, but it can determine only the relative isotopic composition, not the quantities of nuclear materials present.

The decay of the neutron population following irradiations of various lengths was measured for Pu-239, U-238, and U-235 at three electron energies: 7, 8, and 10 MeV. The count rate produced by irradiation with the intense beam from the electron accelerator is high enough to overcome the relatively large steady state count rate due to the Pu-240 spontaneous fissions in the Pu sample. Preliminary analysis shows only slight differences between the delayed neutron decay for a given isotope at 7, 8, and 10 MeV. For the time scale considered, the decay curves are similar to those resulting from fast neutron fission and 15 MeV photofission. At all energies the differences between the various fissile materials are similar to those found in the past from neutron fission. The decay of U-238 is considerably faster than that of U-235, indicating relative richness in fast groups.

One may use any ratio of the delayed neutron population at two different times after the end of irradiation as a signature of isotopic composition. The optimum choice of these times is different for the different materials. With short irradiations one can get a practical 238/235 discrimination ratio based only on the shape of the delayed neutron decay curve of about 2 to 2.5 and with long irradiation this ratio is reduced to about 1.6. Typical discrimination ratios for Pu-239 and U-235 are 1.2 for long irradiations and 1.3 for short irradiations. Although useful for mixtures of U-235 and U-238, these signatures cannot be used efficiently to distinguish between U-235 and Pu-239.

2. Fission Product Gamma Rays

It has been shown experimentally that the fission product yield depends somewhat on the nucleus undergoing fission. Generally, the heavier nucleus has a yield curve which differs by up to two orders of magnitude from that of the lighter nuclide in the mass region A = 100 to A = 120. It is clear that distinctive gamma lines from fission products in this mass region could prove to be a powerful tool in assaying nuclear materials.

In the past, these techniques have been utilized in passive gamma scans, with considerable success, to assay irradiated fuel elements. However, an active interrogation technique using fission products with short half-lives (seconds) has several important practical advantages. For example, it would be very difficult to deceive an active technique that looks at gamma-ray lines with a Ge(Li) detector. This is primarily due to the inherently high resolution capabilities of the Ge(Li) diode. It would be virtually impossible to substitute a material, which upon irradiation, produces gamma lines that have the same energy and relative intensities as those found from irradiating the various fissile materials. Conversely, if one tried to shield for these gamma lines and hide the fissile material, it would be quite apparent in transmission measurements made with the interrogating bremsstrahlung beam.

Several experiments have been performed to evaluate the use of photofission product gamma rays for assay of nuclear materials. A Li-D-Ge detector was used to measure gamma rays induced in 200 gram samples of U-235, Pu-239 and U-238 by 9 MeV bremsstrahlung. From the date obtained, one can draw some immediate qualitative conclusions. One important point is that the intensity of the gamma lines does not seem to be a problem. Reasonably good statistics on a given peak can be obtained in a practical counting time (on the order of a few minutes). Also, the results indicate that one does not necessarily have to have a completely optimized gamma-ray detection system to resolve the lines.

Of course, gamma detection is not limited to active interrogation methods. It may also prove to be very useful for making a passive scan prior to an active interrogation.

3. Threshold Photoneutrons

Experiments are planned to determine the feasibility of using threshold photoneutron measurements as a possible safeguard technique. The threshold photoneutrons with energies roughly from 1 to 100 keV will be detected with a B-10 loaded liquid scintillator. To detect the neutrons in the presence of a gamma-ray background, a pulse shape discrimination circuit will be used at the output of the photomultiplier tube.

The Scanner System

The signatures discussed earlier are presently being studied using small pure isotopic samples, but will soon be employed with bulk material assemblies, such as 55 -gallon scrap barrels and reactor fuel elements. Anticipating the requirements for bulk material measurement, a scanner and detector system has been designed and fabricated. The bulk-sample-scanning mechanism is designed principally for barrel assay work but may also be used for other samples having diameters up to 24 in. It is intended to provide translation and rotation of the sample in order to ensure that the measurement is equally sensitive to every volume element of the sample.

For the 55 -gallon drum assay, the scanning will be performed by exposing an approximately 4-inch vertical section to the incident bremsstrahlung beam. The barrel scan is done in a step-wise vertical movement, with continuous rotation and lateral translation motions. The outputs from position sensors on the lateral translation drive are used to modulate the LINAC trigger pulse rate to obtain a programmed uniform radiation exposure through the bulk sample.

The detector system for the barrel scanner consists of several high efficiency neutron and gamma counters. Two ionization chambers measure incident and transmitted bremsstrahlung radiation, and together determine the material density of the sample.

Analytical Work

1. Precision of Assays Based on an Electron Accelerator Assay System

With present practical techniques, the results of nondestructive nuclear material assays consist of several yields, each of which depends upon the amounts of many or all of the fissionable isotopes present. The error in the result obtained by combination of several yields is not obvious from the yield data, consequently, an analysis has been developed to evaluate the errors in isotopic assays for both real and hypothetical measurements.

The analysis is based on the use of measurements of standard weights of monoisotopes in the same geometry as the measurement of the unknown. A grid search type computer code has heen written which assumes an initial value for the isotopic composition and improves upon this estimate by incrementing the individual amounts one at a time successively. The criterion for improvement is that "chi square" be minimized. The advantage of this grid search procedure over the usual matrix inversion techniques is that non-physical results, such as negative amounts of isotopes, are automatically avoided by the specification of the grid search.

After the solution for the isotopic amounts is found, each amount is changed by increments and the remaining amounts are searched for the minimum chi square. The increment that changes the minimum chi square by one is the precision.

2. Attenuation Effects in the Irradiation of Large Samples

The very poor geometrical conditions encountered in the assay of large samples, such as barrels containing scrap, result in uncertainties in the assay caused by attenuation of the incident photon beam and the emergent neutrons or gamma rays. A new technique has been developed to minimize these effects. The technique depends upon simultaneous rotation and translation of the barrel relative to the bremsstrahlung beam.

In some cases, for which the attenuation of the outgoing products (neutrons) is small, uniform irradiation may be desirable. When the attenuation of the outgoing products is not negligible, it is possible to compensate for it by irradiating the center of the barrel more than the outer regions of the barrel. In this way the yield would be independent of the location of the fissionable material within the barrel. Assuming that the neutron escape probability can be calculated as a function of position in the barrel, the integral equation which gives the required irradiation as a function of position to obtain yields independent of the location of the fissionable material, has been solved.

3. Calculation of the Neutron Yields Expected from Photofission

Since there is little information in the literature on the bremsstrahlung produced photofission yields from nuclear materials, particularly near the fission barrier, a program has been initiated to use simple theoretical models to calculate these yields. A preliminary version of a computer code is now operative.

18. Research Contract No. W-7405-ENG-92 $ 11,000 in FY-68

Title

Laser Microprobe Analyzer

Institution: Battelle Memorial Institute - Columbus

Principal Scientific Investigator: D. F. Askey

Period of Contract: 5/14/68 to 6/30/68

Background & Principle

There are few laboratories which possess the capabilities and technology for determining the isotopic composition of plutonium. The prohibitive cost of mass spectrographic equipment maintained in plutonium facilities has contributed to this problem. However, with the increasing international concern with nuclear weapons proliferation, it would seem desirable to develop a relatively inexpensive method of determining isotopic analyses in plutonium which would be within the budget allowances of all plutonium handling facilities. If this technique can be developed for application to uranium, it can then be extended to the analysis of plutonium.

Objective

Evaluate the use of a laser microprobe to obtain emission spectra for determining the isotopic composition of uranium at reasonable costs. Evaluate the relative sensitivity of this microprobe technique. If the procedure works, it could be extended to refabricated plutonium metal or alloyed fuels. These could be well characterized where the Pu-240 and Pu-241 will be varying by a few hundred relative percent.

Procedure

Fully characterize the emission spectra of plutonium with respect to line intensities and position as a function of isotopic composition as has successfully been accomplished for uranium. The laser microprobe used in conjunction with the emission spectrograph would be investigated as a simple analytical method, especially in view of the fact that the method would be essentially non-destructive, i.e., only a small crater of approximately 50 microns diameter is removed from the sample by the laser beam. No special sample preparation is required; samples of any geometry or form can be readily analyzed.

Analyze metallic uranium samples of varying isotopic composition using laser microprobe, conventional emission spectrograph and time-of-flight mass spectrograph.

Status as of 6/1/68

Experimental work has begun on this limited feasibility study. A summary report will be prepared at the end of the contract period, June 30, 1968.

Status as of 9/15/68

The feasibility of determining the isotopic content of uranium by a laser microprobe-emission spectrograph method was demonstrated. Uranium metal samples of varying isotopic composition from 0.04 to 93 percent U-235 were analyzed by the laser microprobe. Process parameters were defined and working curves correlating spectral-line intensity ratio to U-235 concentration were established for samples with U-235 contents from 12 to 93 percent. Additional experiments performed to define the reproducibility of the data obtained with the relatively unsophisticated apparatus yielded values that were consistent within about ± 5 percent. Further experimental effort leading to application of the technique to plutonium isotopic analysis appears justified.

19. Research Contract No. AT(10-1)-1230

$ 7,000
in FY-68

Title

High-Resolution Gamma Spectrometry for Analysis of Irradiated Fuel

Institution: Idaho Nuclear Corporation

Principal Scientific Investigators: R. L. Heath, J. E. Rein

Period of Contract: 5/2/68 to 6/30/68

Background & Principle

Techniques are needed for the analysis of a variety of reactor fuels in various neutron environments for fissionable material control and reactor usage surveillance in the Safeguards program. The principal techniques which appear promising are high resolution gamma-ray spectrometry for radioactive fission products and isotope dilution mass spectrometry for stable fission products and for uranium, plutonium, and higher atomic number isotopes.

Such techniques will provide the following information: (1) the total number of fissions, (2) the relative number of fissions from the various fissioning isotopes, (3) the average neutron flux of the irradiation, (4) the average neutron energy, (5) the fissionable isotopic content of the preirradiated fuel.

As a nondestructive technique, gamma-ray spectrometry has great promise for many of these measurements and should be optimized for this purpose. Computer-coupled data systems can be developed to apply sophisticated laboratory techniques as the basis for a practical field instrument. Particular emphasis should be placed on the sensitivity of the technique for the measurement of useful fission product nuclide ratios.

The evaluation of the gamma-ray technique may be accomplished by comparison with results obtained from destructive measurements of stable fission products and heavy element isotopes which provide a broader base to obtain the information described above.

The reliability of both nondestructive and destructive techniques depends directly on the reliability of nuclear data for fission yields, capture-to-fission rates (∞), and cross sections. At the present time, the only reliable data available are for well thermalized nuclear reactors. Necessary data must be obtained for typical power reactors operating above the thermal neutron energy level.

Objective

Efforts under this feasibility study are to be limited to the initial design of an irradiation facility and to improving the gamma-ray spectrometry technique.

Procedure

The following work would be included in a complete developmental program (as opposed to the two items listed above for this limited feasibility study).

1. Investigate promising techniques to improve the performance of gamma-ray spectrometers for this purpose. These investigations include the use of larger NaI mantles for the Compton-rejection spectrometer, improving the performance of multi-channel pulse height analyzers at high count rates, and the study of advanced concepts for improved Compton summing spectrometers.

2. Develop computer-coupled data acquisition and analysis systems suitable for practical field instruments.

3. Where reliable data are not available, measure gamma-ray branching ratios and related variables of selected fission products.

4. Irradiate capsules that contain known amounts of the fissionable isotopes in reactor environments typical of the power reactor industry. The objective is to obtain effective data directly applicable to the Safeguards program. The isotopes Th^{232}, U^{233}, U^{235}, U^{238}, Pu^{239}, Pu^{240}, and Pu^{241} will be individually encapsulated and irradiated for various levels of nvt. These levels will be selected to give sufficient fissions to provide reliable measurements of fission yields and as well as to provide adequate information about the heavy element isotopes and the fission product chains and their burnout by parasitic neutron capture and radioactive decay. These irradiations will serve the following functions:

    a. Measurement of absolute fission yields and by the techniques described in the previous section.

    b. Selection of the fission products most useful as monitors of irradiation history and for computation of fissionable material inventory for the various reactor environments.

    c. Provide test samples to aid the development of the gamma-ray spectrometry technique.

5. Because the arrangements for irradiations in operating power reactors and the design and fabrication of the irradiation capsules will require considerable time, preliminary irradiations would be made in the ETR and ATR, extremely high flux reactors. Thermal neutron shields will surround the capsules to simulate power reactor environments. These preliminary irradiations will provide the basis for the selection of the most promising fission products for use as monitors of irradiation history. Based on these data, capsules of selected fission products will be prepared for irradiation along with the fissionable isotopes

in the operating power reactors to obtain effective cross section data directly applicable for Safeguards purposes.

6. Conduct a series of irradiations with typical power reactor fuel samples in operating power reactors. Such samples would be used to prooftest the gamma-ray technique and to obtain working values to "fingerprint" irradiated fuels.

Status as of 7/15/68

Two systems considered to be particularly promising for the measurement of the gamma-ray spectra from irradiated fuels were investigated. These were a DC-coupled amplifier system for improved high counting rate performance of high-resolution spectrometers and an advanced Compton annulus detector for increased spectra resolution. A prototype DC-coupled system was successfully operated that exhibited exceptional resolution performance at rates in excess of 50,000 counts per second. A Compton annulus system was operated with the DC-coupled system in a new coincidence configuration.

20. Research Contract No. $

Title

Isotopic Composition by Delayed Neutrons - Unirradiated Fuel

Institution: Institute für Neutronenphysik und Reactortechnik, Karlsruhe

Principal Scientific Investigator: M. Kuechle

Period of Contract:

Objective

The possibility of determining the isotopic composition and fissile material content in wastes and scraps by the measurement of delayed neutrons is being considered.

21. Research Contract No. AT-33-1-GEN-53 (Related to Safeguards) $ 60,000 to 6/30/68

Title

Gamma Ray Assay of Pu-238 in Waste Drums

Institution: Mound Laboratory, Miamisburg, Ohio

Principal Scientific Investigator: W. W. Strohm

Period of Contract: June 1966 to (continuing)

Results

The amount of plutonium-238 present in contaminated trash and equipment contained in steel drums was determined by measuring, with a NaI(Tl) detector, the intensity of the 765-keV gamma ray from the decay of plutonium-238. By gamma-ray stripping, the contributions of higher energy gamma rays to the region of the 765-keV gamma-ray photoelectric peak could be subtracted, despite the large amount of scattering material present in the drum. The transmission of the 765-keV gamma ray inside the drum was determined by measuring the transmission through the drum of the 765-keV gamma ray from external plutonium-238 standard sources. The uncertainty in the measurements is ± 28% at the 95% confidence level when the drum contains ≥ 0.180 g of plutonium-238.

Publications

W. W. Strohm, "765 keV Gamma-Ray Assay of 238 Pu in Waste Drums", Trans. Am. Nucl. Soc., 10, 41 (1967).
W. W. Strohm, "Gamma-Ray Assay of 238Pu in Waste Drums", Nuclear Applications, Sept., 1968

22. Research Contract No. AT-33-1-GEN-53 (Related to Safeguards) — $ 30,000 to 6/30/68

Title

In-Line Gamma Ray Assay of Pu-238 in Waste Cans

Institution: Mound Laboratory, Miamisburg, Ohio

Principal Scientific Investigators: A. B. Combs, J. Y. Jarvis and W. W. Strohm

Period of Contract: March 1967 to (continuing)

Background & Principle

In part of the plutonium-238 recovery process at Mound, waste contaminated with plutonium-238 is processed in the recovery line and packaged in 0.5-gal cans which are then removed from the recovery line and sealed in #12 cans. The #12 cans are then assayed by the 100-kev Gamma Scan Facility.

The 100-kev gamma scan technique requires that the waste material in the can be categorized, and correction factors are used for each category and for different amounts of plutonium-238 present. Also, this technique cannot accurately measure the amount of plutonium-238 present in cans containing large amounts of gamma-ray absorber.

A technique for assaying the amount of plutonium-238 present in steel drums containing contaminated waste and equipment by measuring the intensity of the 765-kev gamma ray from the decay of plutonium-238 in the drum has been developed at Mound Laboratory and is currently in use. This technique has the advantages of not requiring the waste to be categorized, and the correction for gamma-ray attenuation in the drum is minimized because most gamma-ray mass attenuation coefficients are near their minimum value for 765-kev gamma rays.

Objective

It was desired to significantly reduce the number of cans of nonburnable waste sent back through the recovery line by in-line assay of the 0.5-gal cans before they are sealed in the #12 cans. Those cans containing a significant amount of plutonium-238 can be reprocessed immediately.

Results

An in-line assay technique was developed in which a single-channel analysis is made of the 765-kev gamma-ray photoelectric peak from the decay of plutonium-238 detected by a 3 x 3-in. NaI(Tl) detector.

The single-channel assay technique is suitable for in-line assay and has been installed in the recovery line. The can sits on a turntable in a glove box and the detector is inside a lead shield outside the glove box. The background is extremely high, but preliminary results indicate the technique is satisfactory. However, further data are needed to properly evaluate the technique and to determine if any changes are required.

The procedure was tested by making quantitative single-channel measurements on 17 cans containing known amounts of Pu-238. In order to further develop an accurate 765-kev gamma-ray assay technique, a multichannel analysis of the gamma-ray spectra was made on the same 17 cans.

It was found that the multichannel assay technique is competitive in time and accuracy with the 100-kev gamma-ray assay technique. In fact, a high-energy gamma-ray technique is more accurate for heavy cans than is the 100-kev gamma-ray technique. Furthermore, it is not necessary to know the contents of the can, thus eliminating the necessity of categorizing the cans by type of waste or amount of plutonium-238.

23. Research Contract No. ACDA/ST/RA-20

$ 50,000	$ 55,000
FY-68	FY-69

Title

Safeguards Applications of Precision Mass Spectrometry

Institution: National Bureau of Standards, Washington, D. C.

Principal Scientific Investigator: R. Shields

Period of Contract: 7/1/67 to continuing

Task I. The Application of Mass Spectrometry to Safeguards and to the "Non-Destructive" Analysis of Irradiated Fuel Elements

Procedure

1. Perform high precision mass spectrometric analyses in support of Project MIST (Minor Isotopes Safeguards Techniques).

2. Investigate the possibility of using mass spectrometry as a "non-destructive" method of obtaining the necessary high precision uranium and plutonium isotopic data on irradiated fuel elements using a modern mass spectrometer to perform the analysis on small, representative samples, of the fuel cladding. This type of mass spectrometer is capable of analyzing samples containing less than one microgram of either uranium or plutonium and should be able to provide isotopic data on the micro amounts of heavy elements that have diffused into the fuel cladding during residence in the reactor core.

Status as of August 1968

1. Samples of Yankee reactor irradiated fuel from Cores I through IV have been analyzed for uranium and plutonium. The dissolver solution samples were obtained through the USAEC from the Nuclear Fuel Services chemical processing plant at West Valley, New York.

2. This work is in progress

Task II. Development of Ge(Li) Detectors

Background & Principle

High resolution detectors are needed for the accurate analysis of uranium and plutonium.

Objective

Develop high resolution detectors.

Procedure

Continue the program of basic research on the development of improved, high resolution, solid state, Ge(Li) detectors. These detectors will provide improved instrumentation in several different nuclear safeguard applications.

Status as of August 1968

This work is in progress

24. Research Contract No. $

Title

Plutonium and Uranium Content in Fresh Fuel Elements
Using Slowing-Down Time Spectrometer

Institution: Institut für Neutronenphysik und Reaktortechnique

Principal Scientific Investigator: D. Stegemann

Period of Contract: August 1967 to (continuing)

Background & Principle

It is desirable to improve the analysis of plutonium and uranium by slowing-down time spectrometer techniques.

Objective

Determine the accuracy, time required and the cost of using a slowing-down time spectrometer in assaying plutonium and uranium in fresh fuel pins.

Procedure

The slowing down time spectrometer has been applied for the quantitative and nondestructive determination of fissionable material in fuel pins. In view of nuclear safeguards, three conditions must be fulfilled by such a control method: 1) The total amount of fissionable material within a fuel pin or subassembly has to be determined; 2) a discrimination between different fissionable isotopes, in particular U-235 and Pu-239, must be possible; and 3) the method must be tamperproof, which means in this case that adding absorber materials--in particular resonance absorbers--to simulate a lower content of fissionable material should be detectable.

The slowing down time spectrometer has been assembled and measurements are now being made.

25. Research Contract No. $

Title

Plutonium and Uranium Content in Unirradiated Fuel Pins

Institution: Institut für Angewandte Kernphysik, Karlsruhe

Principal Scientific Investigator: W. Michaelis

Period of Contract: August 1967 to (continuing)

Background & Principle

It is necessary for material assay purposes to develop more accurate methods than are now available.

Objective

Use the different n-γ reactions which are element and isotope specific for determining the plutonium and uranium content with an accuracy of less than 1%.

Procedure

An apparatus has been installed at the FR-2 reactor which consists of a Ge(Li) counter used as a double escape spectrometer and a 44-cm-diam x 60-cm plastic scintillator provided with appropriate wells for the neutron beam and the gamma detector. A lead shield of about 5 cm thickness between sample material and neutron detector prevents the suppression of capture gamma rays coincident with low-energy photons. In principle, all isotopes present in the material including nonfissionable species are detectable.

Measurements are now being made with the equipment.

26. Research Contract No. 121 $ 55,000

Title

Destructive Analysis of Unirradiated and Irradiated Fuels

Institution: Österreichische Studiengesellschaft für
Atomenergie G.m.b.H.
Institute of Chemistry
Reaktorzentrum Seibersdorf, Austria

Principal Scientific Investigator: N. Getoff

Period of Contract: 12/1/61 to 11/30/63

Background & Principle

The purpose of the work was to develop and evaluate analytical procedures for the determination of uranium and plutonium in unirradiated material to ± 1% and in irradiated reactor fuel to ± 3%. Three types of material were to be investigated: (a) uranium oxide fuel; (b) U/Al alloy fuel; and (c) head-end solution.

Procedure

Use of the conventional Jones reductor with ceric sulphate allowed determination of uranium in standard solutions to ± 0.25%. Large amounts of aluminum caused interference and required separating out of the uranium by ion exchange or solvent extraction when the uranium could be determined with better than 0.8% deviation. The Jones reductor is sensitive to zirconium, traces of organic solvent, etc., and furthermore, the necessity of separating out the uranium by ion exchange or solvent extraction is somewhat tedious. Both of these disadvantages were overcome by using chromous chloride instead of the Jones reductor. Combination with a recording potentiometer allowed a further simplification to be made and gave a precision of ± 0.3%.

The methods were tested on unirradiated samples of U/Al-alloy and uranium-oxide fuel and on irradiated uranium from Calder Hall, U. K. In the case of the U/Al-alloy fuel the results were very close to those given by USAEC. The uranium 'dioxide' pellets, on the other hand, appeared to have been oxidized. The plutonium content of the Calder Hall fuel was consistent with the radiation data.

Conclusions

For the determination of plutonium, pre-separation was necessary, and for this, solvent extraction with TTA proved to be the best means. Alpha counting combined with alpha spectrometry and mass spectrometry was used for the determination. The precision was ± 3.0%.

Paper chromatography was shown to be a quick method of separating the uranium from the plutonium.

27. Research Contract No. 314 $ 7,700

Title

Service Contract--Analysis of Irradiated Fuel Plate From BGRR

Institution: Österreichische Studiengesellschaft für Atomenergie
Vienna, Austria

Principal Scientific Investigators: P. Weinzierl, Dr. Bildstein

Period of Contract: 11/9/64 to 2/9/65

Background & Principle

This is a service contract to conduct both destructive and nondestructive analysis on specimens of BGRR fuel.

Procedure

The specimens consist of eighteen pieces of aluminum clad highly enriched uranium alloy having approximate dimensions of 3" X 1½". They were irradiated to a burn-up of about 40% and allowed to cool for a year. The following work was done:

1. Nondestructive determination of Cs-137 content by means of a semi-conductor Compton spectrometer.
2. Determination of uranium content by wet chemistry after dissolution of selected specimens.
3. Determination of the ratio of uranium isotopes by mass spectrometry after chemical separation of fission products from the uranium in the above specimens (Item 2).
4. Determination of the aluminum content in the above specimens (Item 2).
5. Extraction and determination of the plutonium content.

Conclusions

(not available)

D. Gamma Spectrometers

Page

1. Portable Gamma Spectrometer (USAEC) 113

2. Portable Single Channel Gamma Spectrometer (USAEC) 114

3. Development of Safeguards Instruments and Techniques (ACDA) 115

1. Research Contract No. $ 45,000

Title

Portable Gamma Spectrometer

Institution: Argonne National Laboratory

Principal Scientific Investigators: W. J. McGonnagle and N. S. Geyer

Period of Contract: 1959

Conclusions

This program resulted in the development of a transportable gamma spectrometer for use in nondestructive measurements for safeguards control purposes.

2. Research Contract No. AT(45-1)1830 BNW-63 $ 35,000

Title

Portable Single Channel Gamma Spectrometer

Institution: Battelle - Northwest

Principal Scientific Investigator: E. N. Sheen

Period of Contract: 1/1/66 to 12/31/66

Background & Principle

A portable, light-weight gamma spectrometer is required for safeguards inspections.

Objective

Develop a portable light-weight, single channel pulse height analyzer for use as a field instrument to measure the enrichment and quantity of U-235 in "cold" (unirradiated) uranium reactor fuel.

Procedure

Incorporate the following features:

a. scintillation-type gamma spectrometer;

b. compact self-powered unit operating from internal rechargeable batteries;

c. total weight and size to be minimized with emphasis on ease of handling in the field ("suitcase" portability);

d. automatic gain stabilization to be included to compensate for drifts in the high voltage supply, multiplier phototube and amplification system;

e. electronic circuitry to be solid state.

Results

The instrument, contained in two small suitcases, is now being used routinely in safeguards inspections by the USAEC. It is a substantial improvement for this purpose over previously used gamma spectrometers.

3. Research Contract No. ACDA/ST/RA-46 $ 50,000 $ 55,000
FY-68 FY-69

Title

Development of Safeguards Instruments and Techniques

Institution: Naval Research Laboratory, Washington, D. C.

Principal Scientific Investigator: R. Omahundra

Period of Contract: 7/1/67 to 6/30/69

Task I. Gamma Spectral Studies of Enriched US Uranium

Background & Principle

The γ rays from U-235 are relatively weak, 0.19 mev, and are difficult to measure in complex fuel assemblies. However, the high-energy 2.6 mev gamma ray associated with U-232 has been observed in many samples of U. S. enriched uranium now used in research reactors. A knowledge of the initial U-232/U-238 ratio of individual batches of enriched uranium furnished by the fuel fabricator could provide a new method of identifying the material after fabrication and of indirectly confirming the U-235/U-238 ratio for complex power reactor fuel assemblies. It should be noted that the application of this method to an international safeguards system would probably require the use of portable multi-channel gamma ray spectrometers.

Objective

Examine the gamma spectra of many different batches of enriched U. S. uranium to determine if the very small concentrations of the 2.6 mev gamma ray from Tl-208, the daughter of U-232, which have been noticed in some enriched uranium samples, are characteristic and can be used as a diagnostic tracer.

Status as of August 1968

The analyses are now in progress.

Task II. Gamma Spectrometer Development (Multi-Channel)

Background & Principle

Portable instruments are needed to detect the presence, as quantitatively as possible, of uranium and plutonium during safeguard inspections.

Objectives

Determine if existing instruments are satisfactory for safeguards use. If not available, develop as necessary.

Procedure

1. Survey the available commerical portable multichannel γ ray spectrometers and determine if any of these instruments or reasonable modifications of such instruments meet the special requirements for both domestic and international safeguards inspections. If such equipment is not available, design and build a prototype portable multi-channel spectrometer having neutron coincidence detection capabilities.

2. Develop a portable coincidence neutron detector for inspection of plutonium research facilities placed under international safeguards.

Status as of August 1968

Work has proceeded satisfactorily on both 1 and 2 above, and a suitable instrument should be available for field testing during October 1968.

Task III. Non-Destructive Methods of Analysis of Irradiated Fuel Elements.

Background & Principle

Preliminary experiments performed at the Naval Research Laboratory have demonstrated that coincident neutron counting methods can be used to detect the simultaneous emission of neutrons associated with the spontaneous fission of Pu-240.

Objective

Study the coincident neutron techniques as a possible non-destructive analytical method of estimating the amount of plutonium build-up in irradiated reactor fuel.

Status as of August 1968

Work is in progress.

E. Inspection Procedures Page

1. Nuclear Materials Control System (USAEC) 119
2. Formulation and Evaluation of Inspection Procedures for the Application of Safeguards to the Yankee Nuclear Power Station (USAEC) 124
3. Application of Safeguards to Nuclear Fuel Processing Plants (USAEC) 125
4. Application of Safeguards to Fuel Fabricating Plants (USAEC) 128
5. Safeguards Procedures Manual for the NFS Reprocessing Plant (USAEC) 131
6. Development of Safeguards Procedures for Heavy Water Moderated Gas-Cooled Reactor of Pressure Vessel Type With Continuous Refueling (IAEA) 496/RB 133
7. Development of Safeguards Procedures for Heavy-Water Moderated and Cooled Power Reactors with Continuous Refueling (IAEA) 519/RB 135
8. Development of Safeguards Technical Practices for Pressurized Water Power Reactors (IAEA) 577/RO 137
9. Evaluation of Resident Inspection Trial Program (USAEC) 139

1. Research Contract No. AT-11-1-GEN-14 $ 90,000

Title

Nuclear Materials Control System (NMCS) Phase I

Institution: Westinghouse Electric Corporation

Principal Scientific Investigators:

Period of Contract: 12/6/57 to 6/27/58

Background & Principle

A system of procedures and techniques are required by the United States in order to implement the safeguard and control provisions of the Bilateral Agreements for Cooperation between the United States and other governments. Accordingly, the U. S. Atomic Energy Commission in 1957 awarded a contract to the Westinghouse Electric Corporation to conduct a detailed study and develop such a system.

Objective

The purpose of this study was to recommend an effective nuclear materials control system to implement the safeguards on source and special nuclear materials supplied by the United States to other countries, in accordance with applicable agreements for cooperation in peaceful uses of nuclear energy.

Procedure

A specific nuclear complex was assumed for this study as a result of an evaluation of trends in nuclear reactor development. This assumed complex consists of:

1. Four pressurized-water power reactors fueled with slightly enriched uranium dioxide, and producing 2120 Mw of thermal power.
2. Five materials testing reactors fueled with highly enriched uranium and generating 150 Mw of thermal power.
3. A chemical processing plant using the Purex Process and designed for direct maintenance. The annual operating capacity of the plant is assumed to be 75 metric tons of slightly enriched uranium and a few hundred kilograms of high enrichment uranium.
4. A fuel fabrication facility capable of producing the total requirements of slightly enriched uranium dioxide and highly enriched uranium-aluminum fuel assemblies.

The steady-state inventory of SS materials within this total complex is estimated as 140 metric tons of slightly enriched uranium, 400 kilograms of highly enriched uranium and 170 kilograms of plutonium. The value of these nuclear materials is about 50 million dollars, while the capital cost of the nuclear complex may amount to a few hundred million dollars.

It was assumed that the basic requirement of the recommended control system was to insure prompt detection of unauthorized diversion of significant amounts of nuclear materials. Accordingly, critical attention was given to detecting not only the readily conceivable, but to unusual and devious methods and schemes of effecting such diversions. As a consequence, the system recommended incorporates numerous and independent checks to insure its effectiveness. This resulted in a fairly complex system involving the integration of various security and accountability techniques.

Conclusions

A Nuclear Materials Control System (NMCS) is recommended which is designed to implement the safeguard and control provisions of the Bilateral Agreements for Cooperation. The NMCS is intended to detect the possible diversion of significant quantities of source and special (SS) materials from a nuclear complex. It relies primarily on an accountability system for maintaining control over the SS materials. However, completely independent measuring techniques supplemented by physical safeguards are specified because good faith on the part of all operating personnel cannot necessarily be assumed. The recommended NMCS provides "defense in depth" in that multiple checks are available on certain key accountability measurements and several independent protection devices provide physical safeguards for important locations and instrumentation. The majority of the NMCS personnel are stationed at the Central Control Station, which is physically located near the chemical processing plant.

It is believed that the recommended system will amply fulfill the intended requirement, but whether or not the system can be relaxed or simplified appreciably without jeopardizing this function is not known at this stage. Evaluation of this latter possibility will require further development and detailed studies of the system. Thus, the recommended system is believed to be "tight", but it is not necessarily the optimum as regards to simplicity and cost, or attractiveness from the operator's point of view.

Research Contract No. AT(30-1)-2176 $ 1,115,500

Title

Nuclear Materials Control System (NMCS) Phase II

Institution: Westinghouse Electric Corporation

Principal Scientific Investigators:

Period of Contract: 6/17/58 to 6/16/60

Background & Principle

In 1957 the Atomic Energy Commission awarded a contract to the Westinghouse Electric Corporation to conduct an extensive study of the methods of implementing the safeguard and control provisions in the Bilateral Agreements between the United States and other nations. This work, identified as Phase I, resulted in the preparation of a document, "Nuclear Materials Control System, Phase I - Final Report," dated August 15, 1958. This report recommends a system for implementing the safeguards and control provisions in the Bilateral Agreements for Cooperation between the United States and foreign governments. A combination of accountability measurements and physical security controls is proposed. Emphasis has been placed upon the utilization of highly-competent and specifically-trained personnel and special tamper-resistant and fail-safe instrumentation. The system is developed and evaluated in terms of an assumed nuclear complex consisting of power and test reactors, a chemical processing plant, a fuel fabrication facility, and an interplant transportation link.

As a result of the above Phase I work, a Phase II program was initiated for the development, evaluation, testing and tamper-resisting of various instruments, devices, and techniques which might be required in a Nuclear Materials Control System. The Phase II program resulted in the preparation of 33 topical reports which cover the following subjects:

a.	Nondestructive Fuel Element Analysis	4
b.	Nuclear Reactor for Testing Fuel Elements	6
c.	Tamperproof Plant Protective and Monitoring Systems and Devices	9
d.	Computers, Process Instrumentation, Process Analyses	13
e.	Miscellaneous	1
		33

These reports, identified by the letters WCAP- as Westinghouse Commercial Atomic Power department reports, are as follows:

			Page
1.	WCAP-1161	Preliminary Considerations in the Design of the Westinghouse Variable Loading Test Reactor	249
2.	WCAP-1192	The Feasibility of an Annular Core Reactor as a Reactor Fuels Measuring Device	250
3.	WCAP-1221	The Feasibility of an Annular Nuclear Core as a Reactor Fuels Measuring Device	251
4.	WCAP-1232	A Design for the Evaluation of a Reactor Fuels Measurement Facility	252
5.	WCAP-6013	Bilateral Certification of Reactor Design	204
6.	WCAP-6016	Evaluation of Computers for Nuclear Materials Control	202
7.	WCAP-6017	Closed Circuit Television for Security and Surveillance	212
8.	WCAP-6018	Devices and Techniques for Perimeter Protection	213
9.	WCAP-6019	Devices and Techniques for Protection of Enclosed Spaces	214
10.	WCAP-6020	Devices and Techniques for Access Control and Protection	215
11.	WCAP-6021	Devices and Techniques for Protection of Pipe and Conduit	216
12.	WCAP-6022	Alarm Transmitting Systems and the Protection of Communications	217
13.	WCAP-6023	Miscellaneous Devices for Plant Security	218
14.	WCAP-6024	Miscellaneous Devices for Safeguarding a Fuels Reprocessing Plant	219
15.	WCAP-6025	Liquid Samplers for Safeguards Use	261
16.	WCAP-6026	Recording Wattmeters for Safeguards Use	239

17.	WCAP-6027	Neutron Flux Monitors for Safeguards Use	240
18.	WCAP-6028	A Thermal Power Meter for Safeguards Use	241
19.	WCAP-6029	General Activity Detectors for Safeguards Use	220
20.	WCAP-6030	A Gamma-Ray Scanner for Safeguards Use	71
21.	WCAP-6031	Continuous Monitors for Uranium and Plutonium in a Chemical Processing Plant	242
22.	WCAP-6032	Analytical Procedures and Facilities for a Nuclear Materials Control System	12
23.	WCAP-6033	Specific Gravity Instruments for a Nuclear Fuels Reprocessing Plant	243
24.	WCAP-6034	Liquid Level Instruments for a Nuclear Fuels Reprocessing Plant	244
25.	WCAP-6035	Flowmeters for a Nuclear Fuels Reprocessing Plant	245
26.	WCAP-6036	Analyst Performance on Synthetic Purex Solutions	25
27.	WCAP-6038	A Doorway Monitor for Safeguards Use	221
28.	WCAP-6039	The Fuel Assay Scanner	72
29.	WCAP-6040	Experimental Confirmation of the Feasibility of the Variable Loading Test Reactor Concept	253
30.	WCAP-6041	Assay Capabilities of a Reactor Fuels Measuring Facility	254
31.	WCAP-6042	Field Testing of the Fuel Assay Scanner	73
32.	WCAP-6044	A Method for the Nondestructive Analysis of Power Reactor Fuel Elements	74
33.	WCAP-6045	Evaluation of N-16 Monitoring in WTR Primary Coolant for Measurement of Flow and Reactor Power	246

2. Research Contract No. AT(45-1)1830 PNL-7 $ 8,000

Title

Formulation and Evaluation of Inspection Procedures for the Application of Safeguards to the Yankee Nuclear Power Station

Institution: Battelle Northwest

Principal Scientific Investigator: D. P. Granquist

Period of Contract: 1/1/65 to 7/9/65

Results

This contract resulted in the preparation of a report, "Formulation and Evaluation of Inspection Procedures for the Application of Safeguards to the Yankee Nuclear Power Station", (BNWL-119, dated 7/9/65), by the Pacific Northwest Laboratory of Battelle Memorial Institute which records the results of an evaluation of procedures for safeguarding the Yankee facility. The safeguards procedures as defined by the International Atomic Energy Agency were followed. The report summary states:

"A careful evaluation of the safeguards problems in a nuclear reactor area shows that it is primarily a function of exact fuel inventory. Any inspection procedure involving less than full-time resident inspection depends almost entirely upon

a. a positive identification of fuel pieces through the total system, and

b. the absolute containment of neutrons to their intended use within the reactor

Fuel contained within the Yankee reactor is tamper-proof during reactor operation. The positive identification of fuel outside of the reactor requires some additional development effort. Available information shows that it is almost impossible to obtain any appreciable number of neutrons external to the Yankee reactor."

3. Research Contract No. AT(45-1)1830 BNW-70 $ 30,000

Title

Application of Safeguards to Nuclear Fuel Processing Plants

Institution: Battelle - Northwest

Principal Scientific Investigators: D. P. Granquist, R. A. Schneider

Period of Contract: 2/18/66 to 9/1/66

Results

The final report, "Application of Safeguards to Nuclear Fuel Processing Plants" (BNWL-301, of September, 1966) prepared by Battelle-Northwest (Pacific Northwest Laboratory of Battelle Memorial Institute) discusses the procedures required to safeguard a fuel reprocessing plant. An appendix, "A Model Safeguards Laboratory" (BNWL-301 APP) describes a safeguards laboratory, including the costs and manpower required, to be associated with a chemical separations plant. The report summary states:

> "Safeguards procedures to detect possible diversion of nuclear materials to nonpeaceful uses are particularly important in nuclear fuel processing plants because (1) after irradiation, nuclear fuel processing is the first point at which direct physical measurements of a precision consistent with safeguards requirements are possible; (2) any fraction of the total spent reactor fuel which has been dissolved for chemical processing can more easily be diverted without detection than solid fuel; (3) the solutions near the product-end of the plant are sufficiently decontaminated to allow direct rather than shielded (or remote) handling.
>
> "There are, however, some particular difficulties in applying safeguards procedures to chemical plants that process irradiated fuel, for such plants, being designed as they are for maximum flexibility, have both multiple and spare process lines. Thus, the chemical plant operator has at his disposal a large number of flow patterns with which to confuse the inspector. With minor changes, the plant operator could "reroute" material from a normal to a nonnormal exit, and thus "lose" it in some other portion of the plant. He could then recover and process the material whenever the inspector is absent.
>
> "To apply safeguards procedures to chemical processing plants is also difficult because so-called steady-state or continuous plants do not consistently operate at a uniform flow rate. In order that the plant as a whole may stay in balance, subsystem imbalances frequently require that certain sections of the plant operate for short intervals at rates above or below the nominal flowsheet values. Instantantaneous material balances conducted

at such times would be in error unless the accumulated (or depleted) material were taken into account. Therefore an inspection team must have first-hand information about these process variations if it is to adequately interpret material balance or other control data.

"Moreover, after receiving irradiated fuel, a chemical processing plant ultimately dissolves it; and since an inspection team can no longer use piece-count methods to measure throughputs and inventories, it must use a combination of volume and analytical measurements. But the inspection team, when measuring the flow and volume of radioactive process solutions, must rely on instruments that receive signals from remote sensing units; and to calibrate these instruments is difficult. It is even more difficult to be certain that the equipment continues to provide correct readings between calibration checks.

"Because of the particular importance of safeguards in a chemical processing plant and the inherent difficulty of applying them, this report strongly recommends continuous inspection. (It does not seem possible that a few inspectors working a few days a month would obtain enough information to effectively apply safeguards procedures.) This report also recommends that safeguards procedures allow an inspection team to form material balance calculations based on, so far as possible, independently secured measurements, samples, and chemical analyses.

"The effectiveness of a given inspection effort can be predicted only within broad limits. If there is no continuous inspection at the plant, the ability to detect diversion would primarily depend on comparing the product recovered against the calculated input, assuming that the reactor operator is not a party to the diversion activity. Depending upon the circumstances involved, it may be difficult, during a typical 3-month campaign, to determine whether the differences that are less than 10 percent of the throughput are due to diversion or to a combination of poor calculations and process losses. If there is not continuous inspection, the plant operator could easily divert 2 to 5 percent of the throughput without being detected. On the other hand, if the number (and skills) on the inspection team were sufficient to duplicate plant operations, it should be possible to develop an inspection system that could at least duplicate the material control of the plant--typically 1 to 2 percent of throughput during a campaign. But such a large inspection team would unquestionably be too expensive to support, and, in addition, would create intolerable interference problems with plant personnel.

An inspection team whose size is intermediate between these two extremes is limited because it can neither obtain a material balance completely independent of plant measurements and/or sampling procedures, nor observe all actions of every individual in the plant. However, any check on a material balance adequate for safeguards purposes must provide for (1) observing the sampling as well as observing and/or duplicating the analysis of all key samples, (2) observing and/or duplicating the calibration of appropriate instruments and sample tanks, and (3) verifying, through continuous observation, that what is ultimately determined is indeed the true material balance.

"The total manpower required for this intermediate level of inspection will vary, depending upon the size of the facility; its mode of operation; and the degree of independent analytical effort involved. Four to five people will be needed to provide continuous shift coverage at the plant during the processing of safeguarded materials. The need for continuous inspection, for some duplication of analytical effort, and for the accumulation of other independent source data will, for a large separations plant, involve a minimum of about 10 people. About one-half of the required personnel could be junior scientists and engineers or technicians. A sharp decrease in effectiveness is to be expected if the team size is too small. For then, either continuous observation will not be possible or the independent source data obtainable will not be sufficient to verify material balance calculations.

"The major difficulty in forming accurate material balances is that of measuring the irradiated fuel inputs. Further research on methods of measuring, sampling, and analyzing dissolver solution is warranted. A nondestructive method for performing an independent assay would be quite valuable."

4. Research Contract No. AT(45-1)1830 BNW-122 $ 44,000

Title

Application of Safeguards to Fuel Fabricating Plants

Institution: Battelle - Northwest

Principal Scientific Investigators: H. J. Anderson, C. A. Bennett, R. O. Budd, D. P. Granquist

Period of Contract: 7/1/66 to 6/30/67

Background & Principle

There are three main problems in applying safeguards to nuclear fuel-fabricating facilities:

(1) There is the problem of measuring the fuel content of a finished fuel assembly. The measurement uncertainty associated with available nondestructive methods varies widely, depending primarily on the complexity of the fuel assembly. Although a relatively high precision has been achieved in specific instances, the precisions which might be attained under inspection conditions do not appear satisfactory from a safeguards standpoint. Furthermore, in most instances the performance of the assembly in the reactor cannot be used as a basis for estimating fuel content to the required degree of precision. Once the final assembly is fabricated, there is no generally satisfactory nondestructive method of determining its fuel content. Further research and development on the nondestructive assay of nuclear fuel assemblies is needed.

(2) Another problem is the satisfactory evaluation of fuel material contained in scrap. Fuel-fabricating processes typically cause 3 to 20% of the original material to end up as recoverable scrap or disposable waste. The heterogeneous nature of this scrap material has made its measurement, prior to actual reprocessing, notoriously unreliable. Errors of 100% or more are not unknown. In many plants a significant fraction of the recoverable scrap material is accumulated over long periods before the actual material content of the scrap is established. A meaningful material balance cannot be made until good measurements are available for all recoverable scrap and process losses.

(3) Reactor fuel fabrication is typified by specific contracts or campaigns which call for fabricating a definite number of fuel elements or assemblies built to a specific design under rigid specifications. The specified fuel design not only varies widely for different reactors but often varies for the location within the core of a specific reactor. Furthermore, subsequent core loadings for a given reactor often have a completely redesigned fuel. Because of the many different processes and designs which are used in fuel fabrication, it is difficult,

except in a general way, to characterize the process. Thus, a "typical" process yield (in terms of percent of the input fuel material contained in finished fuel elements, recoverable scrap, recycle streams, and losses) may be typical for only one process and fuel design. Therefore, personnel who inspect fuel-fabricating plants must be capable of applying general safeguards procedures and principles to many different fabrication processes and fuel designs.

Objectives

Study the application of safeguards to fuel fabrication plants and prepare a report which will include, among other things, a discussion of;

a. the philosophy of safeguards control in a fabrication plant, i.e., a discussion of the relationship between and relative importance of such factors as inventory verification, nondestructive measurements of cold fuel, and input measurements;

b. the basic rights required, such as access privileges and provision for adequate sampling;

c. any special problem areas associated with fuel fabrication facilities; and

d. the general procedures which would be necessary for the effective inspection of a fuel fabrication plant.

Emphasis will be placed on such items as the necessity for continuous inspection and/or unlimited access, the key points in the process from a safeguards viewpoint, and the relationship of inspection intensity to effectiveness.

Conclusions

Any system of safeguards acts as a deterrent to possible diversion attempts and, if a diversion is attempted, offers the possibility that the diversion will be detected. The amount of material diverted would, of course, affect the probability of detection. An effective safeguards system requires that the inspectors have the right to obtain plant designs, material flows, and suitable accounting records to the extent necessary to account for safeguarded materials. These data provide the starting point for an inspection, or audit, either by resident inspectors or visiting teams.

It is important to recognize that most fuel-fabricating plants are contract-or campaign-oriented. The scrap associated with a given contract or campaign must be processed to the point of obtaining reasonably precise measurements to establish material balances meaningful enough to decide whether or not a diversion has taken place.

Based on this study it is concluded that:

> Depending upon the size of the plant, a resident inspection team of as few as two people, working in a carefully regulated plant, provided with calibrated test standards, and having access to independent, reliable, analytical services, could in most cases achieve a diversion detection capability of about 2% for a completed campaign.
>
> To consistently detect diversions of less than 1% would require continuous intensive inspection of the entire fuel-fabricating process. The improvement from 2 to less than 1% in detection capability is almost entirely due to additional containment and surveillance efforts. This intensive inspection may be difficult to justify both economically and from the standpoint of plant interference.

In a few cases supplemental information may be available to an inspection agency through independent shipper and receiver measurements. Even if these "interface" measurements are assumed to be available, and if the diversion of less than 5% of the plant throughput is of safeguards significance, some degree of resident inspection would appear to be essential for most fabricating plants.

5. Research Contract No. AT(38-1)-452 $ 123,000

Title

Safeguards Procedures Manual for the NFS Reprocessing Plant

Institution: Nuclear Fuel Services, Inc.

Principal Scientific Investigators: R. P. Wischow, J. R. Clark, I. B. Roll

Period of Contract: 6/27/66 to 3/31/67

Background & Principle

The International Atomic Energy Agency (IAEA) is presently verifying the operations of a large number of research reactors and some power reactors, among them the Yankee reactor. Some of the Yankee reactor fuel elements and the contained plutonium are subject to safeguards during the reprocessing operations at NFS. The IAEA safeguards control has been suggested as a mechanism to detect the diversion of nuclear materials to non-peaceful uses.

The present IAEA safeguards system provides principles for safeguards applicable to all parts of the power reactor fuel cycle with specific procedures existing for reactors and for nuclear material located outside principal nuclear facilities. The IAEA Board of Governors established a working group to prepare provisions for chemical processing plant safeguards that are of the same nature as those already established for reactors. The framework of rights of inspectors and the kind of records, reports and inspections which may be carried out are stated but the procedures do not specify a detailed plan for safeguarding a chemical processing plant.

Objective

The objective of this study is to establish a detailed safeguards system for chemical processing plants which will be capable of detecting diversion of nuclear materials if they should occur. A system which has a reasonable expectancy of detecting significant diversions over a short period of time can be expected to be a highly effective deterrent to diversion and in this sense provides an assurance that nuclear materials will not be diverted from peaceful uses. A basic instrument to implement a safeguards program is a safeguards procedures manual.

Results

A system comprised of inspection procedures was developed to allow the NFS Reprocessing Plant to be safeguarded during the processing of safeguarded fuel. The Safeguards Procedures Manual is comprised of three volumes: I Development, II Plant Description and III Procedures.

These inspection procedures were explicitly written for the NFS plant, incorporating the applicable, previously-developed inspection systems and control methods. Application of these procedures to other reprocessing plants of the same general type should be relatively straightforward.

Three types of inspection programs were developed from the procedures; manpower requirements varied from 14-men for a surveillance-only effort, 10-men for the Yankee exercise and 18-men for a program that would approach complete verification of safeguarding fuel to the accountability limits. Estimated annual labor costs for resident inspection are $367,600, $263,600 and $483,600 for the 14, 10 and 18 man programs, respectively. At an annual processing rate of 225 tonnes uranium per year (1,125 Kg plutonium per year) the incremental labor costs for these programs are $0.33, $0.23 and $0.43 per gram of contained plutonium. Costs for the Yankee safeguarding during the processing of fuel is a fraction of the estimated annual costs and is dependent upon the proper requirements developed by the inspection agency.

Safeguarding during processing of Yankee safeguarded fuel can be done with a ten-man team. An intensive on-the-site training is recognized as a prerequisite for this program and a group of observers should be assigned to observe and evaluate the exercise.

Published Reports

TID-23991, "Safeguards Procedures Manual", 3 Volumes, February 8, 1967

6. Research Contract No. 496 $ 19,000

Title

Development of Safeguards Procedures for Heavy Water Moderated Gas-Cooled Reactor of Pressure Vessel Type With Continuous Refuelling

Institution: Power Research Institute, Nuclear Power Research Group, Praha, Czechoslovak Socialist Republic

Principal Scientific Investigator: Milos Drahny

Period of Contract: 12/25/66 to 12/24/67

Principle and Background

The problem is to develop safeguards methods for practical application in respect to natural uranium fueled, heavy water moderated, gas-cooled reactors of pressure vessel type with continuous refuelling. This type of reactor is under construction with supervision of the above Institute. Therefore, the staff of the Institute is the most competent to perform the required work.

To consider the feasibility of use of various techniques and methods for verification of nuclear fuel inventories and operational history of reactor as well as monitoring fuel movements throughout the power plant.

The total cost of the study is estimated as $47,500 of which only $19,000 (40%) is required from the Agency. The rest will be borne by the Institute.

Conclusions

The study resulted in a report which gives an analysis of the possibilities for applying safeguards procedures to nuclear power stations equipped with reactors of the Czechoslovak type (Type A), i.e. heavy-water-moderated, gas-cooled reactors, using natural uranium, with a pressure vessel and continuous refuelling.

A composite method of safeguards control is proposed: the method is suitable for the purpose in question and conforms to the strict requirements of safeguards control.

The proposed composite method consists of 6 operations, which involve the use of partial control methods. Particular use is made of calculations performed by the power station staff for its own purposes: these measurements and calculations are checked by a method independent of the staff.

Although this dividing of the method into 6 operations is regarded by the authors as final, they also suggest another procedure in which partial methods are selected from different alternatives.

Further work on the application of safeguards procedures to Type A nuclear power stations should follow two lines of approach:

(1) Specific development of partial methods, in particular:

automatic counting of the passage of spent fuel elements;

control of fuel elements in long-term storage;

determination of the integrated thermal power of the reactor on the basis of measurements of integrated electric power (this method may also be important for other types of reactor as well as the Type A);

determination of the integrated thermal powers of the separate fuel assemblies and comparison of their sum with the integrated thermal power of the whole reactor;

(2) Selection of a partial method solution for setting up the final composite method; development of this solution for actual use, and economic analysis.

7. Research Contract No. 519 $ 23,000

Title

Development of Safeguards Procedures for Heavy-Water Moderated and Cooled Power Reactors with Continuous Refuelling

Institution: Dilworth, Secord, Meagher and Associates Limited,
Toronto, Canada

Principal Scientific Investigator: F. Hummel

Period of Contract: April 1967 to February 1968

Background & Principle

Safeguards procedures can be broadly defined as means for a country or Agency to ensure that another country is not using special fissionable material to further any military purpose. These procedures are carried out by inspectors given the right of free access to reactor sites under agreement.

The special fissionable materials of principal concern are the fissile isotopes of uranium and plutonium. An inspector would primarily be interested in whether, within a country's work in atomic energy, that State diverted such materials to military purposes.

This study embraces safeguards as applied to the nuclear power station fuelled by natural uranium. The inspection system is required to determine the quantity and isotopic content of plutonium produced by the reactor, and the disposition of this plutonium. Inspection is also required to determine the quantity of incoming fuel. The quality of this fuel does not need monitoring to the extent required in enriched fuel plants. Although the inspector can remain permanently at the plant site, it is naturally preferable that only periodic visits be required, so long as the cost of any systems required to allow periodic surveillance do not outweigh the economics of permanent placement.

Procedure

Specify the procedures required to safeguard heavy water moderated and cooled power reactors with continuous refuelling (CANDU-PHW reactors).

Conclusions

A system was specified for safeguards procedures to be employed in heavy water moderated and cooled power reactors with continuous refuelling. The system employs continuous automatic monitoring, with provisions for selfchecking, to determine whether diversion of fissionable material has occurred from the plant, and also the quantity and quality of such materials held at the site.

In order that a safeguards inspector can cross-check his information, the following three independent procedures should be used:

1. Review operating records
2. Obtain tamper-proof measurements
3. Assay spent fuel

The operating records are used as a basis for the inspector's report. In natural uranium, continuously refuelled reactors a precise calculation of fuel burn-up is carried out continuously by the operating staff.

A concept of tamper-proof measurements has been developed to the stage of feasibility and functional specification. The system monitors irradiated fuel quantity, quality, and destination, by a system of detectors mounted on the fuelling machine, in the fuelling machine vault, and the spent fuel storage bay. The system is simple and centralized, with the output continuously logged by a printer. The system is designed against tampering, and can be made of high reliability. In this regard, all components of lower reliability are made portable and accessible for maintenance and calibration. The system achieves a high degree of accuracy, and will detect abnormal reactor fuelling patterns.

The spent fuel assay system employs a simple method of measuring gamma radiation from a fuel bundle in several pre-selected gamma energy bands to determine burn-up.

The spent fuel assay method determines average exposure power, exposure time and cooling time with no prior knowledge of bundle irradiation history. The method is illustrated by the use of graphs, however, it is anticipated that a form of miniature analog computer would be used to obtain rapid solution. The system would be calibrated by prior gamma measurement of fuel bundles of known irradiation, as such burnup is determined from the repeatability of such gamma measurements. The gamma radiation data used to prove the method was obtained by the use of a digital computer program.

The overall safeguards system proposed minimizes interference with normal plant operations.

The system is applicable to a wide range of reactor types. The spent fuel surveying system can be used on any irradiated fuel, so long an initial enrichment is known. The tamperproof measurement system can be employed with slight modification to any continuously fuelled reactor regardless of cooling cycle, coolant, or moderator.

The system is now at a stage of development where field testing is required on an actual CANDU power plant. This would then be followed by the development of prototype devices.

8. Research Contract No. 577 $ 20,000

Title

Development of Safeguards Technical Practices for Pressurized Water Power Reactors

Institution: J. V. Kurchatov Institute for Atomic Energy
Division of Nuclear Reactors, Moscow, Soviet Union

Principal Scientific Investigator: C. Skvortzov

Period of Contract: 12/15/67 to 6/14/69

Background & Principle

Similar work in the development of safeguards technical practices for heavy water moderated reactors with heavy water cooling is being performed in Canada and for heavy water moderated gas-cooled reactors in CSSR under contracts with IAEA.

The institution has intimate knowledge of the nuclear power plants with pressurized water reactors in respect of their physics, technology design, and operation. The staff of the Institute is the most competent to perform this study. The terms of the proposal, together with additional information supplied by the Institute, satisfy the necessary requirements for safeguards technical development study of this specific type of nuclear power plant.

Objectives

a. To consider the feasibility of using various techniques and methods for verification of nuclear fuel inventories and operational history of pressurized water power reactors as well as for monitoring nuclear fuel movements throughout the plant.

b. To develop practical safeguards methods and procedures for application to pressurized water power reactors with periodic refuelling.

Procedure

1. Conduct studies of the PWR-power plant specifics which are important for safeguards.

2. Make an analysis of fuel movement through the nuclear power plant with PWR.

3. Develop safeguards technical practices for power plant with PWR:

   (a) Plant accountability system to be established for safeguards purposes

   (b) System of reports on reactor operations and fuel inventories;

(c) Detailed inspection procedures;

(d) Possible methods and techniques for use by inspectors.

4. Prepare general conclusions and recommendations for further work.

The programme is expected to be completed in 1½ years. The total cost of the study is estimated at $29,000, of which $20,000 is required from the Agency and $9,000 will be contributed by the Institute.

9. Research Contract No. AT(45-1) 1830 BNW-178 $ 81,000 to 9/30/68

Title

Evaluation of Resident Inspection Trial Program

Institution: Pacific Northwest Laboratory

Principal Scientific Investigator: C. A. Bennett, R. A. Schneider
K. B. Stewart

Period of Contract: 7/10/67 to (continuing)

Background & Principle

With the anticipated growth of nuclear power over the next ten years, a substantial increase in the quantities of special nuclear materials is expected in non-government facilities in the United States and throughout the world. Accordingly, the AEC gave consideration to the need for strengthening domestic safeguards and the procedures that might be used. One of the procedures considered was the use of resident inspection at privately owned facilities, i.e., facilities not under the direct control of the AEC.

Resident inspection in domestic safeguards means the assignment of one or more inspection personnel to a facility for each shift of operation. In general, the function of resident inspection would be to provide for (1) observing the sampling, as well as observing and/or duplicating the analysis of key samples, (2) observing and/or duplicating the calibration of appropriate instruments and sample tanks, and (3) verifying the adequacy of plant operating procedures in validating the material balance source data.

The effectiveness of a safeguards system as a deterrent to, and detector of, the diversion of special nuclear material depends largely on the precision and accuracy with which the quantities of special nuclear material can be measured. However, large quantities of special nuclear material are in such forms or processes that the quantities cannot be measured precisely. In spite of substantial technical progress in measurement methods, it is not possible to eliminate all material unaccounted for and thereby provide 100% assurance that strategic quantities of special nuclear materials could not have been diverted to unauthorized uses.

The essential question is the extent to which resident inspection would contribute to safeguarding of special nuclear material. In an attempt to provide an answer to this question, the AEC has studied the experience of resident inspection programs presently being carried out by the AEC and other government agencies for purposes other than safeguarding material. In addition, an attempt has been made to quantify the additional protection afforded by resident inspection over and above that provided by other material control mechanisms and procedures.

On April 5, 1967, the Commission approved the implementation of a resident inspection program, as an experiment, at the following licensed facilities:

1. Nuclear Fuel Services - chemical processing plant
2. NUMEC - fuel fabrication plant (Pu, U)
3. Nuclear Fuel Services - fuel fabrication plants (Pu, U)
4. United Nuclear Corporation - fuel fabrication plant (U)

Accordingly, resident inspectors (AEC employees) were assigned to the plants for a one year period as follows: NFS-CPP, 10 inspectors; NUMEC-FFP, 1 inspector; NFS-FFP, 1 inspector; UNC-FFP, 1 inspector.

Pacific Northwest Laboratory has been assigned the responsibility of evaluating the resident inspection experiment.

Objective

Establish an initial set of relevant activities for the inspectors which will give the most information about the probability of detecting diversion under a given set of circumstances. Results from these studies, and from the existing body of experience, will be used to build a model descriptive of the system. Output from the model will pinpoint those information gaps that hinder the obtaining good estimates of the probability of detecting diversion. This output will then guide subsequent activities of the inspectors.

Procedure

1. Determine what difficulties are encountered in establishing inspection procedures and plant liaison, and in obtaining necessary knowledge of plant design and practices;
2. List all security situations which will be encountered;
3. For each situation, list the frequency of occurrence in the commercial nuclear industry;
4. List all the possible risks involved, and quantifying these risks when possible;
5. Determining how these risks are affected by the number of inspectors and the degree of surveillance;
6. Suggest remedies in terms of redesign, monitoring, sealing, and containment devices; and
7. Suggest needed research and development work in this area.

Status as of 6/1/68

Detailed information prepared monthly by the resident inspectors at the three fuel fabrication facilities and the chemical processing plant is now being evaluated. The information from the single inspectors at the fuel fabrication facilities indicates that one man cannot possible perform sufficient measurements to arrive at an independent material balance. Accordingly, the assignments of the three individual inspectors have been modified to enable them to acquire data and make enough observations to permit an assessment of the effort and manpower required to make an independent material balance.

The ten man team of resident inspectors at the chemical processing plant has accomplished an independent material balance verification.

This trial resident inspection program is scheduled to terminate at all facilities on June 30, 1968. It is expected that the final evaluation of this experiment will be completed by Pacific Northwest Laboratory by September 1968.

Status as of 10/15/68

A final report is being prepared on the Evaluation of the Resident Inspection Experiment. The experiment was evaluated in the context of two hypothetical safeguards situations under the assumption of attempted diversion. These two situations are:

1. The case of an isolated plant which is not a part of a closed safeguarded measurement system. Under the assumption of attempted diversion, the entire plant force and also any connecting plants in the fuel cycle are regarded as potential adversaries.

2. The case of a plant in a closed safeguarded measurement system where there exists, to some extent, a ring of safeguarded independent measurements for plant inputs and outputs.

The report contains conclusions and recommendations. Among the conclusions are the following:

1. Isolated Plant Concept. To effectively safeguard an isolated plant with current technology, resident inspection is a necessary safeguards activity. It is, however, an expensive undertaking since, to be effective, the inspection force must form its own independent material balance. To form and assure an independent material balance that has a good probability of detecting diversion at a level equivalent to one to two percent of plant thruput, some 10-20 inspectors (depending on the size and complexity of the plant) are required. In addition, an independent measurement capability is required.

For the isolated plant, resident inspection has its highest value. It limits and deters diversion and forces the diverter to take definite risks. In contrast, no inspection opens the door to diversion at a significant level with little fear of detection by normal review or audit techniques.

2. Plant in a Closed Safeguarded Measurement System. In this case, the main safeguards task is that of closing the independent material balance ring. At fabrication-conversion sites, that task is mainly one of verifying waste discards, inventory holdings, and some product shipments. For the fabrication-conversion site studied in the experiment, a small force (1-2 inspectors) can be effective in closing the independent balance ring.

   At a chemical processing plant operating on a campaign basis, there is little inventory between campaigns. As a result, the main safeguards task in closing the independent material balance ring is that of reducing the uncertainty surrounding the independent measurement for plutonium input and verification of waste discards. Because of the large uncertainties currently associated with reactor predicted values for plutonium, the inspection force must independently measure and/or verify the plutonium input. As a result of this consideration and also a need to verify waste discards, a larger force (10 or more) is required at a chemical processing plant to close the balance ring.

F. Inventory Control

Page

1. Inventory Control Study (USAEC) 145
2. Statistical Methods to Evaluate Material Control (USAEC) 146
3. Nuclear Materials Control Manual (USAEC) 150
4. Evaluation of Methods of Input Accountability in a Reprocessing Plant (USAEC) 152
5. Standardization of AEC Inventory Verification Procedures (USAEC) 157
6. Material Accountability in Fluidized Bed Processes (USAEC) 161

1. Research Contract No. $ 123,000

Title

Inventory Control Study

Institution: Vitro Corporation

Principal Scientific Investigators: R. W. Kupp, C. J. Anderson

Period of Contract: 1955-1956

Background & Principle

This study was initiated to investigate the safeguards problems in an international nuclear power complex under the control of the IAEA. The complex consists of ten pressurized water reactors having a total thermal output of 1000 MW, a chemical processing plant, uranium metal reduction plant, and a fuel fabrication plant.

Procedure

The safeguard control schemes considered were: (1) plutonium prediction which is based on reactor power measurements, (2) input measurements which are based on chemical analyses made at the dissolving stage in the chemical plant, and (3) physical security to detect the physical removal of plutonium or uranium.

Conclusions

The study concluded that, following calibration of the reactors, plutonium production can be predicted to an accuracy of ± 2%; the accuracy of input measurements at the chemical processing plant is estimated to be 0.3%. A suitable inventory control system for the reference power complex was estimated to require a staff of 96 people, capital costs of $1,543,500 and annual direct salary costs of $713,000. The cost estimates are based upon 1956 prices.

2. Research Contract No. AT(04-3)-115 $ 54,000

Title

Statistical Methods to Evaluate Material Control (4/66)

Institution: Stanford Research Institute

Principal Scientific Investigators: F. W. Dresch, P. H. Butterfield
E. M. Kinderman

Period of Contract: 9/15/64 to 9/30/66

Background & Principle

The measurement and control of nuclear material has been of concern since 1943-1944 when pilot operations produced the first milligram quantities of enriched uranium and plutonium. In the early history of the varied manufacturing processes, material scarcity was the dominant factor. Today with abundant stocks of most, if not all, special nuclear materials, the U. S. government must apply control procedures to ensure the safekeeping of its large stocks of materials. The nuclear materials industry has grown in magnitude and complexity from its relatively simple beginnings. Operations once conducted in a single government-owned, contractor-operated plant are now often carried out in two or more such plants, in private plants operated for the government, or in private plants operated for private agencies. Industrial organizations, previously without financial responsibility for the materials which they processed, are now fully responsible for large inventories of high specific value materials. Soon, private organizations will own special nuclear materials as well as hold responsibility for them.

Large plant inventories, multiple operations, and private ownership act to increase the complexity of the operation in the nuclear industry, and to emphasize the need for improved management of nuclear materials. The U. S. Atomic Energy Commission holds primary responsibility for the ownership and safekeeping of these materials; the AEC and its principal contractors have developed effective techniques for the necessary measurement and analysis. For a long period of time the AEC has devoted considerable attention to the problem of data interpretation through use of statistical analysis. Enough data and experience have now been accumulated to permit an evaluation with the aim of improving the methods of analysis. Accordingly, the USAEC Division of Nuclear Materials Management awarded a contract to Stanford Research Institute to conduct the evaluation.

Objective

Review current methods and develop improved statistical techniques for monitoring and evaluating shipper-receiver differences, material unaccounted for and normal operating losses. The techniques should indicate if reported differences or losses are realistic and reasonable and should permit the establishment of satisfactory compromise figures.

Conclusions

In the course of this review of present statistical procedures for nuclear materials management within the AEC complex, SRI concluded that the best possibility for improvement appeared to lie in extensive exploitation of system-wide sources of data through integrated analysis. The sophisticated nature of even the most elementary approaches to such analysis, and the wide geographical and organizational distribution of relevant data sources, necessitate organizing on a centralized basis a large portion of the statistical analysis. This would appear to be impractical without computer assistance.

The basic national policy, which has recognized inventories of weapons-grade nuclear materials as a national asset, has given the inventory management problem a different focus than that typical of inventory management problems in industry. Conventional inventory management techniques do appear to be applicable to many parts of the AEC inventory management problem, however, particularly to the nuclear fuel activities. They are also applicable to management of in-process inventories, inventories of raw materials, and production smoothing decisions. Again, the techniques required point to the need for centralized and computerized control for practical implementation of any adequate management system. A centralized computer activity, concerned with system-wide statistical analysis and inventory management, would appear to be a practical solution to both needs.

These inventory and control functions should become parts of the overall information system now being introduced into AEC Headquarters.

The cost of introducing a computerized, statistically-analyzed, control system could amount to $160,000 to $350,000 (including design and implementation of improved data collection and data reporting procedures, computer programming, and pilot testing, but exclusive of equipment and facilities).

This development effort would require about nine man-years of effort over a one-year period. The required force would include two statisticians, one chemist, and one EDP specialist. Operation of the system after shakedown would require a Headquarters staff of three and a cost of about $8,000/month, including computer time.

Introduction of a complete inventory management system would require a staff of 8 to 10 professional operations and systems analysts for 12 to 18 months, with the assistance of AEC staff. The cost of development would be about $300,000 to $600,000 depending on the combination of government and contractor personnel used. A small staff of three to four should be sufficient to run the inventory system after shakedown with a cost of $5,000/month, exclusive of computer time.

Because the statistical analysis and inventory management systems would, to some extent, use common data sources and information storage facilities, savings in both time and cost could be achieved by developing both systems simultaneously, using a coordinated effort.

The coordinated development effort would probably require a staff of 12 to 15 professionals over 12 to 18 months, at a cost of about $400,000 to $800,000.

The number of measurement points and the frequency of measurements, including physical inventories, appear to be adequate but not excessive for the present situation. We believe that the private economy, as it develops, will require more data, especially that relating to shipments and receipts. A move to centralized analysis would require centralized reporting of more detailed measurements, and would eventually suggest the need for additional measurements of specific types--particularly in connection with process models. Some relaxation of requirements for periodicity in inventory measurements could be considered in order to take advantage of naturally advantageous, inventory taking phases in the operating cycles, provided that present average frequency and volumes of data were maintained.

Shipment and receipt measurements are not always made in weapons production chain, and, in some parts of the fuel cycle, are impractical (e.g., scrap shipments). However, adequate control over transfers of materials demands that both shipper and receiver measure the material transferred, especially in the diverse transactions of the private sector of the industry, or between private and government sectors. The measurements must be truly independent ones, reported independently, if statistical or other judgments are to be applied fairly.

Inventory measurements cannot readily be given the same degree of independence, but should be supported by careful calibration and standards programs, and should be checked by the relation of BPIDs and MUFs to S/Rs.

Present procedures for statistical analysis are largely adaptations of quality control charting techniques and are used as trouble warning systems. The techniques themselves, and the manner in which they are used, are generally conservative and traditional, but may suggest trouble more frequently than appropriate or anticipated. This is difficult to assess definitively, since no convenient data bank exists from which overall estimates of biases and measurement precisions can be obtained.

SRI has identified a need for additional staff (primarily statistical) at AEC Headquarters if the present system of control is continued. This staff would consider the more complex statistical problems encountered by nuclear materials management personnel at field offices and Headquarters. As a part of their duties, the AEC staff would develop specialized techniques for statistical analysis similar to those developed by SRI as part of this study.

If the primary recommendation of this report--centralized data reporting and analysis applied to inventory management and materials control--is adopted, a special staff will be required at Headquarters, as discussed above. Some relaxation in staffing requirements at field offices would occur.

Adoption of this report will require a two-stage development program. In the first or study phase, a computer system (software) organization will take the lead role but will work closely with the AEC organization affected. The study phase, lasting three to four months, will delineate clearly the specific approaches to be taken, potential difficulties and advantages, costs, and the preliminary systems concept and specifications for the basic information system and procedures needed for statistical control and inventory management. The AEC, after review of the study phase, would normally proceed with detailed development, programming, and implementation in which it would be assisted by outside organizations, but in which it must play the dominant role. The basic information system needed for monitoring and statistical control of nuclear materials probably could be operable within a year from the start of the study phase. Implementation of more complex and specialized statistical and inventory management techniques would extend over another four to eight months.

3. Research Contract No. AT(45-1)-1350 $ 100,000

Title

Nuclear Materials Control Manual

Institution: General Electric Company

Principal Scientific Investigators:

Period of Contract: 1960 to 1961

Background & Principle

The fundamental basis of any system designed to assure efficient control of nuclear materials is the ability to measure accurately the receipt, issue, use, and inventory of all such materials. Accordingly, the Division of International Affairs of the USAEC awarded a contract to the Hanford Atomic Products Operation of the General Electric Company to prepare four documents which constitute a "Nuclear Materials Control Manual."

Objective

Prepare suitable documents to serve as guidance for organizations setting up a nuclear material control system, primarily for foreign organizations. The documents should present the basic principles and techniques which have been found necessary for the establishment and maintenance of an adequate control system for the nuclear materials used in the United States. In general, the application of these principles and techniques would be required wherever it was necessary for a central agency such as a State to be able to satisfy either itself or another agency or State regarding the safety, national security, and economy of its atomic installations.

Results

The following four documents were prepared:

TID-12099--Nuclear Materials Control in Research and Test Reactor Operations (January 1961)

This report states that the basic unit in a materials control system is the material balance area and then describes the control of material flow and the records and reports required for such control. Various methods of measurement and types of instrumentation are discussed, including the nondestructive assay of highly enriched fuel elements.

TID-12100--Nuclear Materials Control in Irradiated Fuel Reprocessing (January 1961)

The technology of reprocessing irradiated uranium fuel and the measurement technology involved is described. The use of the material balance area and the reports and records required are discussed.

A section is devoted to the audit and inspection of measurement methods, accounting procedures and the use of statistical and mathematical methods for the analysis of data.

TID-12101--Nuclear Materials Control in Reactor Fuel Fabrication
(April 1961)

This document outlines the process technology involved in refining uranium ore, producing uranium fuel, recovering uranium scrap and waste treatment. Measurement technology is reviewed, and the various records and reports to be used in the various material balance areas are described. The final section covers the analysis and control of the system by statistical analysis. This is to assure that the nuclear materials control system permits the acquisition, dissemination and evaluation of quantitative information on the disposition and availability of all nuclear material in the plant.

TID-12102--Nuclear Materials Control in Power Reactor Operations
(May 1961)

The fundamentals of power reactor technology are reviewed. The unique aspects of nuclear material control accounting for reactors includes the calculation of plutonium yields as well as U-235 consumption. The power reactor to which this system would be applied may be fueled with either normal uranium, slightly enriched uranium, or two or more types of fuel in combination.

The records and reports required for use in the various material balance areas are illustrated. In addition, a special internal transfer report form used to record the charging or discharging of fuel into or from the reactor is described.

The report stresses the necessity for analysis and control and states the control system must be continuously evaluated in order to determine whether the system provides the information and control for which it was designed. The results of the evaluation are used to infer either the need for corrective action in order to maintain or restore effective control or the adequacy of, or need for improvement in, the control system itself.

Note:

The above reports are available from the Office of Technical Services, Department of Commerce, Washington 25, D. C.

4. Research Contract No. AT(30-1)-3896 $ 89,000

Title

Evaluation of Methods of Input Accountability in a Reprocessing Plant

Institution: Westinghouse - Nuclear Fuel Division

Principal Scientific Investigators: H. E. Walchli, C. E. Guthrie

Period of Contract: 7/19/67 to 6/30/68

Background & Principle

Irradiated fuel from nuclear power reactors is processed in facilities such as the Nuclear Fuel Services (NFS) Plant, at West Valley, New York. This process separates uranium and plutonium from radioactive fission products and fuel element structural materials. The high value of this fuel necessitates high recoveries and accurate material balances. These requirements make necessary unusually accurate measurements on the plant input, product, and waste streams. The methods used to obtain these measurements and the treatment of the data so obtained define the term accountability.

The present method for input accountability at the NFS plant is based on a tank volume determination and the results of analysis of solution contained within the tank. This approach is based upon several years of operating experience at the AEC owned fuel processing plants.

Recently, methods of analysis have been developed for possible use in alternate techniques for obtaining this input accountability. These alternate techniques would involve calculating the input material quantities, knowing: (1) the pre-irradiation weight and isotopic distribution of the fuel, and (2) sufficient post-irradiation data to enable the calculation of the net change in the quantities of the various isotopes.

At least two possibilities exist for carrying out this calculation. One involves a consideration of the quantity of a particular species of fission product (e.g., neodymium, ruthenium, cesium, etc.) present in the spent fuel; the other utilizes information on the plutonium buildup and uranium depletion in the spent fuel. Although much time and effort has been expended in developing these latter techniques, their application under specific process conditions and an evaluation vis-a-vis the traditional chemical method has not been carried out. One aspect of this proposed program is to conduct this evaluation.

In addition, an entirely different problem arises with the introduction of non-aqueous reprocessing schemes, in particular volatile fluoride. In this process it is not possible (or at least not economically feasible) to use the present chemical methods to obtain feed assay, because of the difficulty in obtaining an accurate volume measurement. Input accountability must be obtained by one of the methods discussed subsequently. The proposed program will provide the background information necessary to assess the accuracy of these methods in instances where the chemical methods cannot be used.

Objectives

a. To provide a sound basis for comparison of the chemical, heavy element isotopic, and neodymium isotopic methods for uranium and plutonium accountability under typical process conditions.

b. To provide an evaluation of the relative accuracy of these methods for plutonium control and antiproliferation purposes.

This program would supplement the analytical samples normally taken by Nuclear Fuel Services, with a second set to be analyzed by an independent laboratory for mass ratios and neodymium and would provide for the further analysis and interpretation of these analyses by experienced Westinghouse personnel.

Provision may also be made for the analysis of the transuranium and transplutonium isotopes of interest by an outside laboratory as well as for U-232 and Pu-236.

Procedure

To obtain the necessary data and establish the format for data analysis the following program will be followed:

a. Prior to dissolution, a schedule will be prepared which will indicate the assembly grouping to be used as dissolver feed.

   Three considerations will be applied: (1) the present location of the fuel in the NFS fuel storage pool; (2) the irradiation history of the fuel; (3) a desire to minimize inter-batch contamination, due to the liquid heel in 3D-1, by having each batch contain approximately the same average burnup. This schedule will be prepared with the cooperation of NFS to minimize the interference with their normal reprocessing routine and to utilize as much as possible their personnel and physical facilities.

   There are potentially four fuel assembly groups to be considered in this program - these corresponding to the first four core discharges from the Yankee reactor. Final assembly groupings will be made when the details for the reprocessing campaign are worked out between the Commission and NFS. Segregation into these groups will assist in the evaluation of the various methods as a function of burnup. To achieve this segregation, the dissolvers will be flushed out between groups.

b. Recognizing that a minimum capability presently exists to conduct the ASTM neodymium analysis, vendor contact will be established early to coordinate the details of the sampling and analysis procedure. The analyses will be contracted on a fixed price basis with one of these laboratories.

c. Samples will be taken from the assay tanks in accord with established NFS procedure. As described in Section II.2 samples are taken from each assay tank. Two are selected for analysis and at least two are set aside for customer or referee uses. For this program, an aliquot will be taken from each of the samples selected for the contract analyses. The volume contained in each aliquot will be ratioed in direct proportion to the volume of the solution in the assay tank from which the original sample was withdrawn. These aliquots will then be composited to make two overall samples; each overall or composite sample will contain one aliquot from every assay tank. The composite samples will be prepared by NFS for the mass spectrometric heavy element and neodymium analysis to be done by the selected outside laboratory. In addition, NFS will do heavy element spectrometric analyses on all composite samples. Each composite sample will be run in duplicate. Based upon a total of four different groups of fuel assemblies to be processed, a total of 16 composite sample analyses will be run at each laboratory.

   Because of the extensive data available on Core I from the Yankee Core Evaluation Program, approximately one-half of the Core I dissolver samples will be submitted to the outside laboratory for heavy element and neodymium analysis. Although these results will not be applicable to an assessment of the material in the complete reprocessing campaign, they will be of value by providing additional data for the inter-method comparison. In particular, the effects of burnup can be investigated in this manner.

   In order to provide a basis for evaluating the statistics of the various methods, it is proposed that monitor and referee samples be included in the analytical scheme. The former would entail resubmittal of a given sample at various times in the program, thus providing data with which the laboratory precision may be determined. The latter would provide for submittal of certain of the samples to the AEC New Brunswick Laboratory for analysis, thus providing a third, objective, check on the overall results.

d. These various analyses will be coordinated and monitored by Westinghouse personnel to assure that the data will be readily amenable to reduction and interpretation.

   In effect five sets of data will be obtained from which the overall accountability can be derived:

   1. NFS contract analyses.

2. Mass spectrometric analyses, performed as part of (1) but analyzed separately by the Heavy Element method.

3. 4. Mass spectrometric analyses on the composite samples, by an outside laboratory, for heavy elements and for neodymium.

5. Mass spectrometric analyses on the composite samples, by NFS, for heavy elements.

In addition, individual inter-method comparisons will be obtained from the detailed analyses of the Core I dissolver batches. Finally, estimates of precision and bias will be obtained from the monitor and referee samples.

e. Reduction and interpretation of the data will entail the following steps:

1. The REBUF code will be modified slightly to handle the neodymium data input and to produce output oriented to this particular purpose. Since this code is now completely developed and operating, the modification required will be small and no difficulties are foreseen.

2. The data reduction will involve processing the data through the REBUF code and applying certain statistical tests. These tests will be devised whereby the three methods of assay (chemical, heavy element mass spectrometric, and neodymium) can be evaluated relative to each other and to the results of the Yankee Core Evaluation Program.

3. The interpretation of the data will be directed toward evaluation of:

   a) Three methods for accuracy and precision;
   b) Relative cost of each method;
   c) Methods as a function of fuel burnup; and
   d) Sensitivity of the plutonium material balance to errors in the various steps in each method.

4. A final topical report will be prepared which will include a complete discussion of the sampling and analytical procedures, the method of data reduction, the interpretation of data, and the conclusions.

Status as of 5/1/68

Eight rerun analyses for uranium composition and concentration were completed by Tracerlab, Inc. during April. The rerun results were in good agreement with the previous values on all but one sample where a transposition of the uranium-233 and uranium-238 intensities measured on the spiked sample had produced gross errors in both composition and concentration.

A more recent submission of the control sample, under another program, showed that the control sample analysis carried out at the same time was low in measured Pu-239/U-238 ratio. Since the lack of any change in the reruns and the unexplained low result on the control sample are in conflict, three samples have been submitted to AVCO Electronics, Tulsa, Oklahoma for analysis for uranium and plutonium.

Nuclear analysis using the heavy element method has been completed for the first 16 dissolver samples using NFS analytical data. Very good agreement exists between the wet chemistry results and the heavy element analysis. However, for the remaining dissolver batches, there is a 1% bias between the results using NFS data. Using composite samples prepared under this program, the uranium agreement is good but a 1% bias on plutonium exists. Resolution of this problem involves additional analytical work and receipt of results from AVCO.

Status as of 10/1/68

A topical report evaluating the results of this program is now being prepared.

5. Research Contract No. AT(45-1)1830 BNW-189 $ 30,000
to 6/30/68

Title

Standardization of AEC Inventory Verification Procedures

Institution: Pacific Northwest Laboratory

Principal Scientific Investigator: C. A. Bennett, R. A. Schneider,
C. G. Hough

Period of Contract: 10/12/67 to 12/31/68

Background & Principle

Preparation of a standardized manual of inventory verification procedures was started by the Division of Nuclear Materials Management of the AEC. Following the reorganization and reassignment of responsibilities for safeguards in the AEC, the new Office of Safeguards and Materials Management assigned to the Pacific Northwest Laboratory of Battelle Memorial Institute the job of completing the manual. This work will involve revising three existing rough draft reports prepared by committees of AEC personnel, expanding on the material as deemed appropriate, and integrating the material into one comprehensive report.

Objective

Produce a finished manual of Inventory Verification Procedures for use in AEC surveys of SS material holdings. The ultimate objective of the effort is to provide certification statements associated with the determination of SS material inventories resulting from AEC's surveys of SS material holdings. The manual is intended to develop predetermined protection levels designed to improve AEC-adopted safeguards goals.

Procedure

The effort will consist of the following steps:

1. Review the three rough draft reports constituting the framework upon which the manual will be based;

2. Study of the comments provided by the AEC field offices on these reports;

3. Revise the rough draft reports in view of the comments in 2., and other considerations;

4. Prepare a final manual on "Standardization of AEC Inventory Verification Procedures."

Status as of 5/1/68

The "Manual for Inventory Verification" has been organized into four parts:

Part I.	The Measuring Process
Part II.	The Sampling and Stratification of Materials
Part III.	Sampling Plans for the Verification of Inventory Procedures
Part IV.	Sampling Plans for the Verification of Inventory Quantities

Efforts to date have been confined to the conceptual design of the manual content and to obtaining actual data from facilities throughout the country that can be used as examples in the manual. Processing of available data is progressing well. However, much of the type of data needed for Part I does not appear to be readily available. Therefore, it may be necessary to use some examples from existing literature sources in order to complete the manual within the intended time period.

The manual as now planned includes some additions to the Subcommittee Reports on Stratification, Statistical Aspects and Sampling Procedures.

The Subcommittee report on Statistical Aspects will be substantially unchanged in content. Organization of the material is revised in order 1) to emphasize that this section (Part III) is primarily concerned with verification of the procedures used in taking an inventory (as opposed to verification of quantities of material that make up an inventory), and 2) to distinguish between verification of measurement procedures (weight, volume, sampling and analytical) and verification of procedures for the recording of inventory transactions.

The Subcommittee report on Sampling will be expanded considerably in Part II to include statistical developments in the field of bulk sampling of materials. The problems associated with sampling of materials are intimately related to the problem of stratification of materials. Thus, the Subcommittee report on Stratification will also be expanded in Part II to show the interaction of physical measurements of materials and the sampling of materials as they affect stratification of these same materials.

This effort goes farther than originally planned by the Subcommittee on Stratification and is not necessarily a prerequisite to the statistical aspects of verification of inventory procedures. However, it is considered an important prerequisite to the statistical aspects of the verification of inventory quantities.

The verification of inventory quantities also goes beyond the original goals of all three committees. It is considered that extension of present inventory verification practice to include verification of inventory quantities is essential to realization of effective nuclear material safeguards. Accordingly, the proposed outline includes two new sections (Part I, IV) that were not included in the original manual concept.

Part I organizes and defines measurement of material properties as a "measurement process" that is imposed on the materials in a physical process. Part I also defines and organizes the statistical methods as they apply to the measurement process. Thus, statistics is here used as a tool in the study of the "science of measurement" and the "science of materials." Part I will only be an introduction to the subject but will indicate the need for continued activity in this most important area of nuclear material safeguards.

Part IV is an extension of the verification of inventory procedures to verification of inventory quantities. As in Part I, Part IV will be introductory in nature and will discuss concepts that are not fully developed.

Status as of 9/30/68

As a result of the initial work on this project, it was determined that the manual should be extended to include the verification of total amounts of material and estimates of the associated measurement bias and precision.

Inventory survey data obtained from several contractors has been analyzed. These data analyses serve two purposes. One purpose is to provide case studies (examples) for inclusion in the manual. A more important purpose is to learn the statistical properties of the data so that correct statistical procedures can be designed which will ensure verification of total amounts and provide certification statements that satisfy pre-determined protection levels.

These data analyses have strongly confirmed that detection and virtual elimination of defective measurements is essential to verification of total amounts of material. However, the accepting or rejecting of an inventory based on the observed number of defective measurements (items) is, of itself, not sufficient to ensure verification of total amounts of material. The study indicates that in some cases an observed proportion defective of 0.125 (12.5%) has a minor effect on the total amount of material. Other cases, where the observed proportion defective was 0.02 to 0.04, can be shown to have a major effect on the total amount of material. These facts have lead to the development of procedures to determine the error introduced by defects and to factor the magnitude of the error into decisions for accepting or rejecting a lot. There still remains the problem of deciding whether the observed measurement bias and precision (after disposition of error introduced by defective items) meet safeguards criteria for verification of total amounts of material. Thus, certification statements based upon pre-determined protection levels for verification of total amounts of material are dependent upon at least five factors: 1) proportion defective, 2) error introduced by defective measurements, 3) measurement bias, and 4) measurement precision, and 5) propagation of error over various strata to arrive at the precision and bias associated with total amounts in inventory. Current efforts are aimed at designing the statistical procedures that will enable the incorporation of these factors into decisions concerning the disposition of a lot.

Effort has also been aimed at evaluating various (five) sampling plans that are currently used in the field to accept or reject a lot based on the observed number of defective measurements in a sample of items. All of these plans have one thing in common: they are approximations to the hypergeometric distribution. As a result, the relationships between lot size and the sample size required to ensure a pre-determined protection level are different, and in some cases, are poor approximations to the hypergeometric distribution (small lot sizes). Current efforts are aimed at utilization of existing sample plans prepared by the Sandia Corporation that are based on the exact hypergeometric distribution. Adaptation of these plans may enable replacement of the five "approximate" sample plans by one set of sample plans that are based on the exact distribution. The result would be a set of graphs (OC curves) and tables for sample size that would eliminate complex computations and the inconsistencies of the approximate plans.

Although the manual will be different in content than originally planned, it will have little effect on actual field practices. Its greatest effect will be on the data analysis and computational phase which is done after the actual plant survey. The goal is to develop sound statistical procedures that reflect the realities of a complex measurement system. Such procedures will form the nucleous of contractor efforts to improve the measurement system and meet safeguards criteria, both domestic and international.

A technical report describing procedures for analysis of weight data for proportion defective and measurement precision and bias has been completed. Work is proceeding on the evaluation of numerous sample plans and on the selection of specific plans that are compatible with the principles of analysis demonstrated in the Weight Data Report.

6. Research Contract No. AT (10-1)-1230 $ 35,000
to 9/30/68

Title

Material Accountability in Fluidized Bed Processes

Institution: Idaho Nuclear Corporation

Principal Scientific Investigators: D. R. DeBoisblanc, J. A. Buckham

Period of Contract: 7/1/67 to 9/30/68

Background & Principle

Fluidized bed processes for recovering uranium values from expended reactor fuel elements are now undergoing pilot plant investigation and may soon be expected to advance to plant-scale recovery operations. Examples include burn-leach or combustion-dissolution processes for graphite-containing fuels and halide-volatility processes for metallic fuels. In order to control such processes from both material accountability and safety standpoints, reliable sampling techniques will be vital. At present, no proved sampling devices are available for operation in beds of fluidized solids of varying size, density, and physical form.

AEC installations, as well as future commercial fuel processors, will be using fluidized bed processes of diverse nature, and the body of information developed in this program should prove invaluable in design of accountability systems for such processes. The Office of Safeguards and Materials Management should find the results of the investigation valuable in evaluating the measurement programs of future processors using fluidized solids processes.

Objective

To study and evaluate methods of achieving material accountability in fluidized bed processes: specifically, to study determination of material quantity and solids sampling techniques in equipment containing fluidized beds of non-homogenous solids.

Procedure

Accountability studies will be conducted in actual processing vessels in the course of the ICPP pilot plant investigations and thus will not involve simulated situations or special construction to provide a testing environment. It is proposed to extend the study beyond the bare minimum necessary for design of the specific system to be used in the ICPP processes. By extending the experiments to include evaluation of samples withdrawn from various locations in different vessel configurations and different process conditions, as well as different sampling equipment designs, it is expected that criteria can be developed that will permit a reasonable evaluation of sample variations due to sampler design and installation in other fluidized bed applications.

Status as of 6/1/68

Experiments with the fluidized-bed graphite-fuel burner indicate that wall effects apparently influence the composition of samples taken from a fluidized bed through a circular opening in the vessel wall. Fine particles of graphite which were known to be in the bed, and passed overhead with elutriated product, were found in greatly reduced quantity in samples withdrawn through a 3/8-inch diameter sample line tapped into the vessel wall and finished smooth at the inner surface.

Other preliminary experiments indicate some tendency toward size segregation with height of bed material in a fluidized bed system and, with uranium oxide coated on alumina, a possible increased concentration of uranium at the top of the bed where the particles are smaller, but presumably have a higher particle density.

During a study of heat transfer in fluidized beds of solids with varying particle size distributions, a series of samples were taken for study in the accountability program. Samples were withdrawn from the fluidized beds at different bed depths using sample outlets at the vessel wall. The size distributions of particles in these samples will be compared with those of the whole beds, determined both before and after the test program. Representative beds used in the heat transfer study will be saved for possible further experiments in the accountability program, depending on the outcome of the current sampling study.

A pilot plant run of several days duration is being planned in the graphite fuels fluidized bed burner. A complete accountability study and materials balance determination will be made in connection with this run which will be conducted with uranium-containing fuel.

G. Mathematical Models for Fuel Cycle Analysis

Page

1. Mathematical Models and Decision Functions Study (USAEC) 165
2. Systems Studies for Special Nuclear Material Safeguards (USAEC) 167
3. Nuclear Process Analysis Studies (USAEC) 171
4. Mathematical Model of Fuel Cycle (BMwF) 176
5. Projekt Spaltstoffflusskontrolle (BMwF) 178
6. Fissile Material Flow Control at Strategic Points in a Reprocessing Plant (BMwF) 179
7. Use of Statistical Analysis for the Establishment of Material Balance in a Reprocessing Plant (BMwF) 180

1. Research Contract No. AT-(04-3)-165 $ 100,000

Title

Mathematical Models and Decision Functions Study

Institution: Thompson-Ramo-Wooldridge

Principal Scientific Investigator: D. L. Drukey

Period of Contract: 1960

Results

This study by Thompson-Ramo-Wooldridge resulted in the preparation of a final report, "Final Report Under Phase III of the Nuclear Materials Control System", which consists of two volumes.

Volume I

The operations in a typical Nuclear Complex such as might be safeguarded overseas were analyzed and methods were derived for utilizing the results of measurements made in the Complex to insure that the nuclear materials had not been lost or diverted. The methods may be extended to a variety of nuclear facilities in addition to the specific complex studied.

The Nuclear Complex used as a model for this analysis consists of nine reactors, a chemical reprocessing plant, and fuel fabrication facilities to support the reactors. There are five MTR-type reactors, two Yankee-type and two Dresden-type reactors. The chemical reprocessing plant provides the capability for processing approximately 100 tons per year of uranium from discharged reactor fuel elements and for recovering the associated plutonium. The fuel fabrication plant consists of three lines; one for highly enriched MTR-type fuel, one for low enrichment PWR and BWR-type fuels and one scrap recovery line.

Measurements are made at principal locations in the Nuclear Complex at varying time intervals to determine the location and quantity of U-235 and plutonium within the Complex and to determine that none have been lost.

The decision functions which have been derived under this contract represent mathematical expressions into which the results of the measurements may be inserted to determine whether or not a loss has occurred. The requirement for somewhat sophisticated expressions of this kind arises because the basic measurement data are not absolutely precise. Each measurement is subject to a limitation of precision inherent in the technique of sampling and analysis. The objective of the decision functions is to make the best possible decision, taking into account the limitations of precision of the measurements which make up the material from which the decision must be formulated. The decision functions presented here are dependent on the attainable precision of the measurements. For this analysis, precisions are used which are attainable with techniques which are currently in use or in proven laboratory status. The considerations which go into establishing the decision functions involve the susceptibility of the decision system to false alarm (that is, indicating that a loss has occurred when such is not, indeed, the case) and the sensitivity of the detection system to an actual loss when it does occur.

Volume II

The task reported here consisted of analyzing a typical Nuclear Complex such as might be operated as a semi-self-contained unit in an overseas area. The analysis aims to develop inventory policies appropriate to economic operation of the entire complex. It permits planning for the amounts of uranium and plutonium to be maintained in inventory in various parts of the complex and provides typical reorder policies for key elements of the complex.

The economic model derived determines the inventory levels which are to be maintained at various points within the complex and the bases upon which orders for shipments are placed. Shipments are scheduled to maintain a roughly constant flow of material to the processing plant. When the inventory behavior throughout the system to meet material demands is understood, a more satisfactory and efficient control model may be developed.

In summary, if actual material inventories vary greatly from those calculated to be economically optimum, or if a reactor or chemical reprocessing plant is operated on a schedule which is obviously far from optimum, there would be cause for suspecting that nuclear materials were being used or produced for improper purposes.

2. Research Contract No. AT(49-2)-1165 NBS Project No. 4314429 — $105,000 to 9/30/68

Title

Systems Studies for Special Nuclear Material Safeguards

Institution: National Bureau of Standards

Principal Scientific Investigator: A. R. Vincent

Period of Contract: 2/1/68 to (continuing)

Background & Principle

In formulating bases for future standards and criteria of performance for safeguards, it is important to distinguish capability from performance. Performance requirements for a new plant should reflect both capability and the extent which that capability has been achieved in practice. This is true for both process capability and the ability to measure and account for nuclear materials.

The National Bureau of Standards (NBS) has been chosen to carry out that portion of the system development where, for the most part, contact must be made with licensees. The result of this work will be the development of a mathematical model which will simulate the nuclear fuel cycle.

To reach the immediate goal of deriving bases for standards and criteria of performance, the most readily available experience data will be examined by NBS. The task is centered on determining historical yields, measured losses, material unaccounted for, BPID, shipper-receiver differences, and measurement uncertainties. From these data, recommendations for interim standards and criteria of performance will be derived. The current plan is to emphasize fuel fabrication processes while assigning a lower priority to chemical processing plants. The AEC has an immediate need to develop standards for fabrication and material conversion sites while the next commercial chemical processing plant is several years away.

The Nuclear Process Model will be structured basically along the lines of a material flow model assuming, initially, that the only uncertainties involved are those due to measurement and location. Once the general mathematical framework of the model is structured, then restrictions or weighting factors can be introduced to take into account the fact that the source data and flow information may be deliberately altered to mask diversion. In general, these weighting factors will include degree of independence of the measurements and accounting functions, the extent of surveillance, physical containment, security, etc. that assure the credibility of source data and material flows.

Objective

The objective of this program is to develop system models for nuclear facilities which can be used to evaluate alternative safeguards systems from a material accountability point of view. One of the basic questions to be explored is how the uncertainty of specific measurements of the amount of material flowing through or within a system (i.e., a nuclear facility) influences the total uncertainty for the system. The results of these studies are intended to identify areas where better measurement techniques would cause a significant reduction in the total uncertainty and to establish a material accountability standard for the various processes within a nuclear facility.

Procedure

The problem areas dictated by the objective of this program are concerned primarily with determining the effectiveness of nuclear safeguards within a nuclear facility. Emphasis will be placed on the collection, the analysis, and the interpretation of data relative to the amount of material flowing through various unit processes and on identifying those areas where data is needed and not available. The output from each unit process and/or material balance area will be partitioned into the desired product, the measured operating loss, the material unaccounted for and the amount of scrap generated. Associated with each of these determinations is a measurement uncertainty, therefore, studies will be made to characterize these uncertainties and to determine the total uncertainty for the system. Periodic visits will be made to nuclear facilities to gain a detailed understanding of various processes, measurement techniques, inventory levels, and sampling procedures. If suitable data are available, a material accountability and control model will be developed for a particular plant. It is expected that such a model will provide a rationale for standards and specifications for the operations of several nuclear facilities.

Status as of 7/1/68

Visits were made to the Hematite, Missouri, plant of United Nuclear Corporation and to the Erwin, Tennessee, plant of Nuclear Fuels Services. Data covering the conversion process, $UF_6 \rightarrow UF_4 \rightarrow U$, were collected at both plants. Material measuring points were identified, and the precision of the measurements was determined. The data are now being analyzed to establish how they can best be used to develop a mathematical model of the process. Based on the data, the process parameters which influence material unaccounted for (MUF) and Normal Operating Loss (NOL) will be identified.

It was observed that although the equipment and process methods used at the two plants are quite different, the material inputs and outputs are the same. Based on this information, a study is being made to determine the feasibility of developing a mathematical model for the $UF_6 \rightarrow UF_4 \rightarrow U$ conversion process. The model will indicate how well material can be accounted for in terms of the equipment used, the size of the job, the enrichment and the measurement system.

Detailed information was obtained at the NFS Erwin plant on the following processes:

1. $UF_6 \rightarrow UO_2$
2. $UF_6 \rightarrow U$
3. Pelletizing; U-233 O_2 - ThO_2
4. Pelletizing; PuO_2 - UO_2
5. Scrap Recovery (above 5% enrichment)

Process data covering the past 18 months were obtained for $UF_6 \rightarrow U$ metal conversion on enrichments above 5%.

Data obtained at the UNC Hematite plant covered the following processes for highly enriched uranium:

1. $UF_6 \rightarrow UF_4$
2. $UF_4 \rightarrow$ U metal
3. $UF_4 \rightarrow UO_2$
4. Scrap Recovery

Status as of 9/12/68

An accountability model has been developed and programmed for a computer which is capable of assessing, quantitatively, the MUF and its uncertainty. This uncertainty of MUF is derived in terms of the uncertainties associated with applicable measurements made in a process or collection of processes. The model was developed with a background of information obtained from visits to several nuclear facilities. This information was obtained by tracing the flow of nuclear material through all stages of a specific process, identifying where nuclear material is measured, estimating the precision of these measurements, and collecting historical data on performance of various processes. This information can be used for predicting the range of MUF's one would normally expect as well as the associated frequencies of occurrence.

Concurrent with the above development, the analysis of historical data on the performance of a process is being considered as a means of measuring process effectiveness. This information can be used to establish normal losses across various subprocesses, determine in-process holdup, and provide criteria for performance limits in future facilities. Also, it can be used to signal when a process or a collection of processes requires attention from a safeguards point of view.

The accountability model when coupled with a model characterizing performance is capable of assessing quantitatively any changes in the safeguards system due to the introduction of different measurement devices or techniques. It will show how each individual measurement influences the total uncertainty of the system. A significant feature here is that Cost-Effectiveness studies can be made for alternate measurement systems.

3. Research Contract No. AT(45-1)-1830 BNW-192 $ 91,000
to 9/30/68

Title

Nuclear Process Analysis Studies

Institution: Pacific Northwest Laboratory

Principal Scientific Investigators: C. A. Bennett, R. A. Schneider

Period of Contract: 1/29/68 to (continuing)

Background & Principle

The development of an effective overall safeguards system requires the performance of nuclear process analyses with the short-range goal being the development of criteria for the safeguards system. The long-range goal of the studies is the establishment of a Nuclear Process Model which incorporates all the parameters pertinent to the overall safeguards system. The key items are unit process stages and flows, measurement uncertainties, degree of independence, historical indices of performance, site-to-site couplings, storage, measurement costs, etc. It is envisioned that the model will be used as a research tool to:

1. Simulate new processes and measurement systems to predict the expected yields, losses, indices of safeguards performance, and the probability of detecting diversion if attempted.

2. Derive cost-effectiveness relationships.

3. Derive bases for standards and specifications for new processes and revise bases for old ones in view of improvements in the safeguards system.

4. Simulate the effects of newly evolved safeguards technology to predict the effect on costs and safeguards effectiveness.

5. Indicate weak spots in the safeguards system and pinpoint areas for further R&D efforts and indicate those areas where R&D efforts would not be fruitful.

The nuclear process data is being derived from two main sources: First, process and measurement capabilities; and, second, operational and accountability experience.

Processing capabilities are considered to consist of material flows, yields, losses, and inventory levels which are dictated by the nature and capability of the process and for which yield and loss data are based on long-term experience. The term "process experience" as used here denotes historically demonstrated process capability rather than performance. Process yields and losses refer to yields based on measured processing losses and nominal thruput and not based on operational upsets and unmeasured losses such as leaks, losses to the atmosphere, undissolved solids, etc. Measurement capabilities refer to the capability to measure the amounts and flow (or location) of the materials.

Operational and accountability experience reflects both operating performance and the experience acquired in measuring and accounting for the material. Key data include total measured losses; unaccounted for losses; inventory uncertainties and their influence on BPID; shipper-receiver differences; sampling, analytical and bulk measurement experience; and associated measurement costs. The sum total of capability and performance data make up what is termed nuclear process experience.

Battelle Northwest (BNW), which operates Pacific Northwest Laboratory (PNL), has been assigned the additional task, under its present contract, to perform nuclear process analyses primarily among the AEC cost-type contractors.

Objectives

The objectives of the safeguards research and development program are to assist in the achievement and maintenance of an effective safeguards program, both domestically and internationally. These objectives will be attained by (1) improving present safeguards systems and techniques, and (2) developing new systems and techniques applicable to the expanding nuclear industry.

The safeguards systems studies are currently concerned with the analysis and evaluation of processes and operations in the nuclear fuel cycle, the mechanisms by which scientific methods and techniques are applied to safeguarding special nuclear material, and the handling of the data which results from such applications.

Projected system studies, or more appropriately, operations research, must expedite the determination and evaluation of plant capability based on theoretical studies and plant performance based on historical data. The operations research will provide information from which the AEC can develop standards and criteria for normal operating losses (NOL) and materials unaccounted for (MUF).

The requirements of physical security during process, storage or transit cannot be ignored, whether in terms of personnel surveillance or by dependable electromechanical devices. A simple, depersonalized well-instrumented system is desired.

Procedure

1. Perform detailed systematic studies of accountability experience and processing experience of AEC contractor fabrication processes. This effort will be minimal until the immediate goal of interim standards and criteria is met for fabrication processes.

2. Formulate a mathematical Nuclear Process Model.

Status as of 6/1/68

Criteria and Standards for Safeguards Performance Indices

Conceptual approaches and guidelines are being developed for the establishment of criteria and standards of performance for the following indices:

1. Normal operational losses (NOL)

2. Accidental losses (ALL)

3. Material unaccounted for (MUF)

4. Shipper-Receiver Differences (S/R)

Current efforts center on describing and determining the basic components of each index of performance. Particular emphasis is being given to relating this work to the fundamental material controls (FMC's) described in the "Proposed Guide for Preparation of Fundamental Materials Controls and Nuclear Materials Safeguards Procedures" Guidelines and bases for establishing interim criteria and standards for material conversion and fuel fabrication processes are being developed.

Nuclear Process Simulation

The nuclear process model has been extended to include all the measurement uncertainties associated with a material conversion process. The uncertainties due to measurement imprecision and biases are propagated for each working unit and variances are summed over all working units to obtain estimates of the overall uncertainty of the book inventory and the material balance index, MUF.

The computer-based model has been generalized to simulate multiple nuclear processes. The model can now act as a framework on which fixed calculation routines such as Burnup calculations can be added at a later date. The model has been generalized in the sense that the working units will be described by input parameters rather than fixed logic in the computer. As a result, the flow of specified amounts of material can be simulated through the entire cycle and the uncertainties surrounding the material at any point in the cycle can be described.

Status as of 9/30/68

Computer Phase of the Nuclear Process Simulator

The computer-based process simulator is currently programmed in FORTRAN for the Univac 1108. For small simulations only core memory is required. However, as larger systems are simulated, secondary storage will be needed in the form of storage drums. For the initial phases of the simulation studies, information is feed to the computer via punched cards.

The flow of material through the model can be represented in the following ways:

1. Entirely by discrete measured quantities at all steps in the process.
2. By a combination of discrete measured quantities and a fractional flow matrix.
3. By a combination of nominal batch sizes and rates and a fractional flow matrix.

Discrete measured quantities and/or nominal batch sizes are used to specify input to the process model from an outside working unit, for ending physical inventory, and for the transfer of discrete quantities of material from one working unit to another or to another site. Where discrete quantities are not measured, the flow of material is represented by a fractional flow matrix. The row of the matrix indicates the location from which the material is being transferred and the column indicates the location to which the material is transferred. The magnitude of the fraction indicates the fraction of material which is recycled to some earlier process stage.

In operation, the simulator consists of three functional components - an input component, a material processing component, and an evaluating component. The input component specifies the process flow parameters, the quantities per batch, and the measurement errors associated with each batch and each measurement point within each working unit. A number of these interconnected working units comprise the material processing component. Each working unit operates on a quantity of input material, splits it into product and various types of scrap, waste, and recycle fractions, and sends the resulting output quantities to other working units. All or any fraction of the input quantity may be retained in the working unit to represent an ending inventory. From the data specified by the input component, the material processing component simulates the material flows and at some specified time computes the indices of performance (S/R and MUF) and their associated uncertainties. The evaluation component allocates the total uncertainty to the various working units and measurement points and computes the ability to detect a specified level of diversion.

The simulator is now being tested on material conversion processes.

Flow sheets have been completed for the power fuel cycle, and cost-effectiveness studies are in progress.

Error Propagation Model For Material Balance Simulator

A mathematical model for individually observed measurements has been developed in which explicit formulae are given for computing MUF and V (MUF) when single or multiple working unit material balance areas are used. The accountability item is uranium weight as determined by the product of process unit weight and analytical fraction U. Gaussian propagation of error is used, assuming independence of all quantities considered. Although in a particular practical situation this independence assumption may be unfounded, some insight into the basic problems and capabilities of material balance accounting as a safeguards tool can be pointed out using this simple approach. The process simulation studies are designed to determine the effect of various non-independence situations on material control as one of its outputs, so that this problem is ignored at this time only for the sake of simplicity in presenting some broad considerations. No novel statistical concepts are presented.

The basic conclusion is not novel either. It is that as the size of the material balance area increases, internal transfers are lost to control by the indices MUF and V(MUF) as these indices become more a function of inventory difference. Thus, the indices provided by shipper-receiver differences, S/R, and V(S/R) become more important in controlling the uncertainty surrounding the material in the system.

4. Research Contract No. $

Title

Mathematical Model of Fuel Cycle

Institution: Institute für Angewandte Reaktorphysik, Karlsruhe

Principal Scientific Investigators: W. Gmelin, D. Gupta, W. Häfele

Period of Contract: March 1967 to (continuing)

Background & Principle

In order to establish a safeguards system which will serve to prevent or detect the diversion of nuclear material, the entire nuclear fuel cycle must be examined to determine where material can be diverted. Also, it is necessary to know what is the least amount of diverted material that can be detected.

Objective

Conduct preliminary studies preparatory to a systems analysis of the nuclear fuel cycle.

Results

A report was prepared titled, "On Modern Safeguard in the Field of Peaceful Application of Nuclear Energy, " (KFK-800, May 1968). The report is divided into two sections:

I. Basic Considerations

II. Preparational Considerations for a System Analysis

In summary, this report concludes that the application of safeguards consists of three safeguard measures;

(1) Containment; nuclear material enters a facility through only one entrance and leaves only through one exit. This principle must be extended to transportation and to control of the waste stream.

(2) Strategic Point Control; control, by sampling and analysis, the flow of nuclear material in each facility of the entire fuel cycle.

(3) Inventory Control; measure the material hold-up in a facility by a physical inventory or by a plant clean-out.

For an effective safeguards system, the efficiency must be quantified and a criterion specified by the safeguarding authorities such as, "The requirements of safeguard are met if with (95)% confidence level the material balance is closed within (2)%." The exact figures must be developed by a detailed systems analysis.

The identification of strategic points of the fuel cycle and the evaluation of their efficiency is the first task in the design of a safeguards system; it should be possible to do this within a year or two. Consideration must also be given to establishing ground rules for nuclear facility design for safeguards purposes. Finally, tamperproof process measuring instruments and automatic data processing systems must be developed.

Part II of this report develops preparational aspects of the mathematical analysis of the safeguards problem.

5. Research Contract No. $

Title

Projekt Spaltstoffflusskontrolle

Institution: Institut für Angewandte Reaktorphysic, Karlsruhe

Principal Scientific Investigator: D. Gupta

Period of Contract: December 1967 to (continuing)

Background & Principle

In preparation for a broad program to develop equipment and procedures for safeguards, a detailed worklist and five year time schedule, "Vorstudie zum Projekt Spaltstoffflusskontrolle," has been completed. (February 13, 1968) Included in the outline report are estimates of financial and personnel requirements.

The program consists of the following subprojects:

1. System Analysis
2. Indirect Methods (Non-destructive analysis)
3. Direct Methods (Destructive analysis)
4. Tamperproof Devices and Procedures
5. Fuel Cycle Model
6. Assembly of Safeguards System
7. Cooperation with German Industry and Government
8. International Cooperation

6. Research Contract No. $

Title

Fissile Material Flow Control at Strategic Points in a Reprocessing Plant

Institution: Kernforschungszentrum Karlsruhe
Federal Republic of Germany

Principal Scientific Investigators: A. von Baeckmann, W. Gmelin, D. Gupta, W. Häfele

Period of Contract: March 1967 to (continuing)

Background & Principle

The main purpose of a fissile material safeguards system in a fuel reprocessing facility is to prevent or detect any diversion of fissile material from the plant. Of the three basic safeguards measures, namely, containment, material balance and other redundant methods, available to a control authority, the second measure plays a key role in a properly developed safeguards system. A complete containment of fissile material may not be economically practicable in a reprocessing plant operating industrially, and an excessive use of the third measure may require undesirable intrusion in the plant or impose a heavy burden of activity on the control authority. Whereas a material balance for a plant can be established relatively economically and without causing an excessive intrusion into the plant or an excessive workload on the control authority.

Conclusions

This study resulted in the preparation of a paper, "Fissile Material Flow Control at Strategic Points in a Reprocessing Plant," (KFK 801). The conclusions reported are:

With the help of a fissile material balance around a reprocessing plant, it is possible to assess the probability of a diversion after it has taken place. This means there is always a finite time lag between the time of diversion and its detection, and a diversion cannot be prevented directly by carrying out a material balance alone. However, with a proper choice of strategic points in a plant and a combination of other safeguards measures, the material balance may decrease the possibility of diversion in an indirect manner and may also reduce the time lag between a diversion and its detection.

7. Research Contract No. $

Title

Use of Statistical Analysis for the Establishment of Material Balance in a Reprocessing Plant

Institution: Kernforschungszentrum Karlsruhe
Federal Republic of Germany

Principal Scientific Investigators: W. Gmelin, D. Gupta, W. Häfele

Period of Contract: March 1967 to continuing

Background & Principle

In carrying out fissile material balance in a reprocessing plant, considerable amounts of information and data are required. Since the accumulation and use of these data may cause a heavy burden of work on the control authority, it is desirable to find out ways and means in reducing the burden of work.

Results

This study resulted in the preparation of a paper, "Use of Statistical Analysis for the Establishment of Material Balance in a Reprocessing Plant," (KFK 802).

In this paper two possibilities of reduction of the work burden have been discussed and analyzed. The first possibility deals with compositing individual samples obtained from tanks or containers at different strategic points for the determination of fissile material content. The second possibility deals with the use of data obtained by the operator himself for the determination of fissile material balance in the plant. As illustrations for the two methods, numerical examples have been given for the data obtained from a fuel element storage area. The high plutonium content fuel forms the basis of the calculation.

Conclusions

On the basis of the study, the following conclusions can be drawn:

The operator's data can be utilized by the controller for establishing his own material balance provided the following three conditions are fulfilled:

(a) The data of the operator have to be controled by the reliance of the first* and the second** type.

(b) The required standard deviation for the test has to be guaranteed.

(c) It has to be clarified what type of measure should be taken in case the data from the operator significantly deviate from those of the controller.

In case these conditions are fulfilled the utilization of operator's data offers the following advantages over the case in which completely independent measurements are carried out by the controller for the establishment of the material balance.

(i) The effort required for carrying out chemical analyses is considerably less.

(ii) In case no significant deviation exists between the operator's and the controller's data, the use of the former improves the absolute standard deviation in kg Pu/day of the controller's measurement compared to an independent material balance by the single sampling method. This improvement is about 5% in the case of the composite sampling method and about 30% in the case of the single sampling method if the operator's data are utilized. But the analytical effort required for the composite sampling method using the operator's data will be only 6.7% of the effort required for an independent measurement. For a single sampling method the effort will be the same.

(iii) The composite sampling method shows some disadvantages if used by the controller for the establishment of an independent material balance. This method can however be utilized in combination with operator's data effectively because the values of possible falsification in the operator's data can be regulated by a proper choice of the statistical confidence.

*Reliance of the first type is based upon the probability with which falsified measurements can be discovered.

**Reliance of the second type depends upon the statistical confidence interval for the test between the operator's and the controller's data.

H. Miscellaneous

	Page
1. Technical Support Organization (USAEC)	185
2. Minor Isotopes Safeguards Techniques (ACDA)	191
3. Development of a Photographic Identification System for Fuel Elements (IAEA) 244/RB	193
4. Investigation of the Radiation Chemistry of Plutonium Nitrate Solutions (USAEC)	195
5. Fuel Sample Stability (USAEC)	199
6. Evaluation of Computers for Nuclear Materials Control (USAEC)	202
7. Radioactive Waste Containers for Storage and Shipping (USAEC)	203
8. Bilateral Certification of Reactor Design (USAEC)	204

1. Research Contract No. AT-30-2-GEN-16 $ 134,000 to 9/30/68

Title

Technical Support Organization

Institution: Brookhaven National Laboratory

Principal Scientific Investigator: H. Kouts, W. A. Higinbotham

Period of Contract: 1/19/68 to (continuing)

Background & Principle

At Information Meeting 762 on January 19, 1968, the Commission approved the establishment of a Technical Support Organization (TSO) at the Brookhaven National Laboratory as set forth in Staff Paper AEC 132/124 dated January 10, 1968.

Objectives

The functions of the TSO as set forth in Appendix A of AEC 132/124 are as follows:

FUNCTIONS OF A SAFEGUARDS AND MATERIALS MANAGEMENT TECHNICAL SUPPORT ORGANIZATION

I. (a) Analyze data from systems studies of national and international, contractor and licensee fuel and material conversion and fabrication cycles performed as a part of the safeguards research and development program to provide technical advice to the AEC regarding possible changes in methods, techniques and procedures to reduce Normal Operational Losses (NOL), Materials Unaccounted For (MUF), and scrap inventory.

(b) Provide technical assistance for AEC development of standards and criteria for safeguards systems.

II. As authorized by the Office of Safeguards and Materials Management (OSMM), perform or have performed technical studies needed to maintain and improve an adequate safeguards program. Provide technical advice and guidance to the AEC:

(a) for areas where additional study is needed;

(b) for discontinuing studies that are not making, or do not appear to be capable of making, a useful contribution to the safeguards effort; and

(c) regarding the value of proposed research and development projects.

III. Based on the analyses performed in I and the application of research and development performed in II, propose suggested instrumentation, independent verification techniques, and other scientific methods and procedures to improve and simplify safeguards and safeguards techniques. The estimated costs as a result of the proposed suggestions would be evaluated.

IV. Examine data generated by a computer based data evaluation system to provide the AEC with an evaluation of the effectiveness of the safeguards system.

V. Through continuing review, evaluation, experiments, and studies, call attention to areas in the safeguards system that may be susceptible to diversion and as authorized, develop or have developed instrumentation, measurements, and controls to reduce or eliminate this susceptibility.

VI. Through continuing review and evaluation of the nuclear industry growth and growth predictions, provide technical advice and guidance to the AEC regarding modifications of the safeguards program needed to keep pace with the industry.

Procedure

The Technical Support Organization will work on the following specific tasks:

1. Study of the "Strategic Point" concept of safeguards
2. Review of Research and Development Proposals
3. Review of Research and Development Projects
4. Scrap Measurement Technology
5. Investigate Applications of Portable Gamma Spectrometers to Non-Destructive Assay Problems
6. Use of Seals as a Safeguards Tool
7. U. S. Safeguards System Definition and Description
8. Parametric Analysis of Nuclear Process Models
9. IAEA Staffing for Safeguards Under the Non-Proliferation Treaty
10. Glossary of Terms
11. Plant Instrumentation Field Tests

Status as of 6/1/68

Work is now in progress on all of the specific tasks. Effort to date has included much of a preparational and organizational nature. As results are obtained and conclusions reached, either topical or progress reports will be prepared for each task.

Status as of 10/1/68

The progress of work on the 11 specific tasks is as follows:

1. Study of the "Strategic Point" Concept of Safeguards

The strategic point concept has been proposed by the Federal Republic of Germany as a specific method to safeguard nuclear facilities. TSO has studied this approach and compared it with the material balance concept currently employed in the United States. Both methods involve measurement of the materials flowing into and out of a plant or port of a plant in order to assure that all material is accounted for.

As research and application of the different methods continues, the philosophies and techniques become more clearly defined. Similarities and differences are being clarified so that factors such as cost, effectiveness, credibility and political acceptability may be compared. There would appear to be little difference of opinion with regard to the location of measurements at strategic points within the fuel cycle. Further discussion and analysis will be required to develop a universally acceptable approach to the roles of physical containment and surveillance activities.

2. Review of Research and Development Proposals

OSMM receives many proposals for research and development projects related to safeguards. These proposals are reviewed by the OSMM staff. They are also reviewed by TSO for technical feasibility, relative importance to the safeguards program and to determine whether the level of effort and proposed program is likely to obtain the desired results within a reasonable period of time. TSO has also analyzed the current research program and the proposals and may solicite proposals for areas which appear now to be neglected.

3. Review of Research and Development Projects

OSMM has in progress, and will have in the future, many safeguards research and development projects. The OSMM staff maintains close contact with these projects and progress thereof. There is a need, however, for independent evaluation of the projects as they progress, are completed, and are followed by other projects, to maintain a coordinated pattern of research and to optimize R&D spending.

TSO has collected information on American instrumentation and R&D which may be of interest for safeguards purposes and has begun to exchange such information with the U. S. Arms Control and Disarmament

Agency, the International Atomic Energy Agency and with institutions in other nations which have active safeguards programs. The contractors supported by OSMM have been visited by TSO personnel, as well as a number of other institutions which have projects of interest. On the basis of these discussions and an evaluation of the current status of instrumentation and data handling in the United States, recommendations have been prepared for research and development to be conducted in the next few years with special emphasis on techniques and experiments which promise early results.

4. Scrap Measurement Technology

Several typical nuclear facilities were visited, and a paper was written which describes the various types of scrap and waste products produced and the types of containers used to store them. Methods currently being used for scrap analysis, both destructive and nondestructive, are being surveyed. Some of the instrumentation being developed and described elsewhere in this report is specifically intended for scrap analysis.

5. Investigate Applications of Portable Gamma Spectrometers to Nondestructive Assay Problems

The sodium iodide scintillation detectors and single channel analyzers now used by U. S. and IAEA inspectors were evaluated. A number of samples of uranium and plutonium were analyzed using high-resolution germanium detectors and multichannel analyzers. In cooperation with the Arms Control and Disarmament Agency, several portable passive analysis instruments are being procured for evaluation in the field. Information has been collected from a number of institutions on the latest techniques in high resolution gamma ray spectroscopy and passive neutron measurements which may be useful now for national and international inspectors or field tests of safeguards systems.

6. Use of Seals as a Safeguards Tool

Two related programs are underway. One is an analysis of the use of seals: when may they be used to advantage and what function may they be expected to perform. The other is to collect information on the types of seals which are now available and to evaluate their performance. TSO is working with the International Atomic Energy Agency to find out how difficult it may be to open and reclose the seals now used by the Agency in such a way that the operation would go undetected. This investigation may lead to a conclusion that the seals are adequate for their purpose, that they may require some modification or that the technique needs revision.

7. U. S. Safeguards System Definition and Description

TSO has been collecting information on the present U. S. safeguards system in order to relate the research and development program to the current needs. The systems analysis studies described elsewhere will also contribute to this objective. The U. S. safeguards system is continually being reviewed and revised. The outcome of these studies will be embodied in a report which describes the present and projected U. S. safeguards program in a comprehensive manner.

8. Parametric Analysis of Nuclear Process Models

Nuclear process models are being developed, as described elsewhere, at the National Bureau of Standards and the Battelle Northwest Laboratory. The design and use of these models will be coordinated to provide maximum usefulness to OSMM in developing its understanding of safeguards systems and planning for the future. TSO will make use of these studies to identify the more sensitive points in the nuclear fuel cycle, to suggest what areas of research and development deserve special emphasis and to design meaningful field experiments.

9. IAEA Staffing for Safeguards Under the Non-Proliferation Treaty

The first report written on this heading was presented to the Committee on Foreign Relations of the U. S. Senate, July 17, 1968.* This is a forecast of the costs for IAEA safeguards for the period 1970-1990 based on assumptions which are compatible with the current IAEA system as presently defined. As the IAEA system develops, its performance and costs are clearly of great interest to all participating nations. TSO will continue to study the growth of the nuclear industry and the safeguards systems, staffing and costs in cooperation with all other interested parties.

*(Hearings on the Non-Proliferation Treaty, before the Committee on Foreign Relations, U. S. Senate, July 10, 11, 12, 17, 1968, pp.153-155, 281-288.)

10. Glossary of Terms

A glossary is being prepared of the special terms currently being used in the nuclear safeguards field. Definitions are being collected from many sources, edited, rewritten and circulated for comments to interested parties, both in the United States and abroad.

11. Plant Instrumentation Field Test

One conclusion of a recently completed study was that safeguards R&D should be oriented toward developing the more sophisticated techniques and procedures that will be needed in the near future. One factor in these more sophisticated, or higher levels, of safeguards is the depersonalization of the safeguards system by the use of plant instrumentation to replace, or at least minimize, the need for resident inspection personnel. The system studies presently in progress will identify critical points in the nuclear fuel cycle that will require surveillance either by instrument or personnel. There are, however, many such points that can be and have been identified on the basis of past experience and evaluation of currently available data. Investigation of instrumentation at these points is now in progress and is being coordinated with the results of the systems studies as they are obtained and with the current technical studies which are investigating instrument development.

This study will evaluate available instrumentation, determine what development is needed and obtain and field test selected instruments. The field tests will measure the effectiveness of instruments in providing safeguards information and will provide an evaluation of their cost-effectiveness for safeguards.

2. Research Contract No. ACDA/ST-140 $ 25,000 FY68-69

Title

Minor Isotopes Safeguards Techniques (MIST)

Institution: Battelle Memorial Institute, Columbus, Ohio

Principal Scientific Investigator: R. Ewing

Period of Contract: 7/1/67 to 6/30/68

Background & Principle

The minor isotopes safeguards techniques (MIST) project is based on the use of the ratios of the minor uranium and plutonium isotopes to the major isotopes. The present concern for developing safeguards procedure for the power reactor fuel cycle suggests an initial investigation of these ratios as means of identifying and tagging nuclear material as it proceeds through fuel fabrication, reactor irridation and chemical reprocessing. Project studies are being conducted at the Nuclear Fuel Services, West Valley Plant, the National Bureau of Standards, and the Battelle Memorial Institute in Columbus, Ohio.

Objectives

Assemble the isotopic data on uranium and plutonium and determine if the minor isotope ratios can be used to safeguard and characterize the fissionable material in such nuclear facilities as reactors, fuel reprocessing and fuel fabrication plants.

Procedure

Use the ratios of the abundance of minor isotopes of uranium U-234 and U-236 to the abundance of the major uranium isotopes U-235 and U-238 to accomplish the following:

1. Characterize or "finger print" individual or specific batches of uranium.
2. Identify the processes that have altered the isotopic composition of the uranium, for example: (a) enrichment, (b) irradiation.
3. Verify that certain industrial operations have been performed as specified, for example:
   A. The blending or mixing associated with reactor fuel element fabrication.
   B. The desolution associated with the chemical processing of irradiated reactor fuels.

C. The recovery of fissionable scrap and waste materials.

4. Monitor overall the operation of certain major nuclear facilities, for example:

    A. Reactor Operations.

    B. Chemical separation plants

It should be noted that many of the applications of MIST cited above are also directly applicable to plutonium using similar ratios of the minor isotope Pu-241 to the more abundant plutonium isotopes Pu-239 and Pu-240.

The major premise underlying the application of MIST is that the latent diagnostic information contained in the abundance of the natural isotopic tracer U-234 found in all uranium samples, and the artificial isotopic tracer U-236 found in those samples that contain at least some uranium which has been exposed to neutron irradiation, should be used to the maximum extent possible.

Status as of 9-30-68

The initial report on fuel from the Yankee reactor is expected during October, 1968.

3. Research Contract No. 244 $ 50,000

Title

Development of a Photographic Identification System for Fuel Elements

Institution: Stanford Research Institute
California, U.S.A.

Principal Scientific Investigator: E. M. Kinderman

Period of Contract: 12/15/63 to 6/30/65

Background & Principle

Accurate inventory of special nuclear materials is required because of their high specific value and for safeguards purposes. Special nuclear materials in the form of fuel elements and fuel element assemblies are among the easiest to identify and control.

Visual observation and identification is the simplest of all techniques. Photographic inspection is nearly as simple and in addition has the value of permanent recording. It permits, for example, comparison of markings recorded at two stages in the fuel element history: at final assembly in the fuel manufacturers' plants and in the irradiated fuel storage pool at the utility power plant.

The first of these stages (the fuel element manufacturers' plants) offers no difficulty or hazard to the person seeking to photograph some particular and characteristic marking. (This could be a lot or piece number, an inspector's stamp, or a weld with characteristic appearance.) The second stage requires some radiation shielding between the material and inspector. Very commonly this shielding is the water in a fuel element storage pool. Photographic recording under water shielding or in high radiation fields seems more difficult than in the first instance because of the distance between the object and observer and the poor illumination and optical distortion caused by the shielding media (water, lead glass).

Under this contract an investigation was made of the techniques and procedures required for photographing objects under water while retaining the high resolution necessary for unequivocal identification. Also investigated were techniques for preserving any marking placed on the element or assembly during its manufacture and the ability to record identification markings photographically at other stages of the fuel cycle.

Procedures

To provide a system for positive identification of reactor discharged fuel, a compact and convenient optical examination system utilizing a fixed examination station, a periscope, a telescope, and a camera was developed. The system was tested in a cobalt irradiation pool by examination of test coupons which were marked in various ways, and then treated in a hot, circulating water loop. In the test system, a water-filled periscope, 11 feet long, a catadioptric telescope placed 16 feet from the periscope, and a 35 mm camera for recording the observations were used. The telescope, camera, and mountings are all readily portable for they weigh in total less than 22 pounds and have maximum dimensions of 29 inches.

Photographic observations of test resolution charts in air 20 feet away from the object lens of the telescope have demonstrated that the system can resolve markings 22 microns in width. This corresponds to a resolution of 0.8 second of arc. Tests performed on the water system have shown that resolution in this system is better than 50 microns at 20 feet. This corresponds to a resolution of about 1 second of arc. It is clear that the resolution can never be better than the air path system. The present system used in an actual fuel inventory should reproduce any inventory markings.

Conclusions

A photo-visual inspection device was designed, constructed and tested. It was found that such a system will permit identification and detection of irregularities in reactor fuel inventories. Of course, the periscope is only necessary for those reactors using water as a loading and storage media. With moderate care one will be able to standardize the photograph size by adjustment of the target-objective lens distance to have the same effective path length for successive photographs. This would be desirable in the case where stero projection of the negatives is used to detect forgeries.

The telescope system has a resolution of about 0.8 second of arc. It can be reproduced at a cost of about $2000, and is easily transported in normal air travel.

It is recommended that the photo-visual inspection device be used in conjunction with stereographic projection of the negatives (control and subsequent negatives) for inspection programs. It may be desirable to have a high magnification ocular lens. This, together with visual information obtained by the inspector, can prove to be a reliable means of unequivocal identification.

It is further recommended that the improved system be tested under actual operating conditions at a reactor during a safeguards inspection.

It will be necessary for each reactor facility to have a periscope, and therefore, we recommend that the periscope design be given separate consideration.

4. Research Contract No. AT(29-1)-1106 $ 97,000
to 10/31/68

Title

Investigation of the Radiation Chemistry of Plutonium Nitrate Solutions

Institution: Dow Chemical Company, Rocky Flats Division

Principal Scientific Investigator: F. J. Miner

Period of Contract: 7/1/67 to 10/31/68

Background & Principle

Nitrate solutions are used in all of the major irradiated fuel reprocessing plants in this country (Hanford, Savannah River, and Nuclear Fuel Services) and abroad (Windscale in the United Kingdom, Mol in Belgium, CNEN-PCUT in Italy). In this reprocessing, nitrate solutions are used in the initial dissolution of the fuel elements as well as in the subsequent separation of plutonium, uranium, and fission products from each other.

During dissolution of the irradiated fuel element, the solution is exposed to intense alpha, beta, and gamma radiation. Reactions involving plutonium which are induced by this irradiation must be understood because the species of plutonium present could be altered in such a way that normal sampling and analytical measuring techniques would give erroneous results.

In the separation of plutonium from the other components of the dissolver solution, plutonium is commonly produced in the form of a nitrate solution. These solutions are subjected to intense alpha radiation by the plutonium. As in the dissolver solutions, reactions induced by this irradiation must be thoroughly understood so that proper sampling and analytical techniques can be used to accurately determine the plutonium content.

The Purex Process (the major process used in this country for reprocessing fuel elements) usually includes an evaporation step to concentrate the final plutonium nitrate solution for shipping or further processing. This concentration step oxidizes part of the plutonium to Pu(VI). Previous work has shown that with Pu(VI) present there can be effervescence and even precipitate formation in the solution. This continues until Pu(VI) is eliminated. The effervescence and precipitate formation lead to solution instability and can cause problems in storing, shipping, sampling, and analyzing the plutonium nitrate solutions.

Objective

The objective of this proposed investigation is to develop criteria for solution composition of plutonium nitrate solutions that will lead to the stablest solutions possible under the conditions imposed by the fuel reprocessing methods.

Procedure

The criteria will be developed by:

a. Investigating the effects of alpha, beta, and gamma radiation on the chemistry of plutonium in nitric acid solutions so as to provide an understanding of the chemical behavior of plutonium in these solutions, and by

b. Investigating the effect of the hexavalent oxidation state of plutonium on the stability of plutonium nitrate solutions.

Status as of 3/1/68

Gamma Radiolysis

An experimental plan has been developed to determine the effect of NO_3 concentration, H+ concentration, temperature, and O_2 (air) on the gamma radiolysis products of HNO_3. Using a 5000 curie Co-60 source, the concentration of gaseous products H_2, O_2, N_2 and nitrogen oxides were found to be in general agreement with the limited amount of comparable data available in the literature.

Beta Radiolysis

A beta irradiator consisting of a 500 curie Sr-90 source was obtained. Following the design and installation of a suitable sample solution circulating system, the collection of dose rate data was initiated.

Alpha Radiolysis

Arrangements have been made to obtain PuO_2 - 238 microspheres to be used as an alpha source. Handling procedures have been specified, and shipping arrangements have been completed.

Methods of Analysis

Accurate analyses of the various radiolysis products are required for this program. Conventional spectrophotometric analytical methods were evaluated for NO_2 and H_2O_2 and were modified as required for use in HNO_3 solutions.

As soon as sufficient data has been developed on the effects of alpha, beta and gamma radiation on HNO_3 solutions, of varying concentrations, similiar solutions with plutonium added will be investigated.

Status as of 9/1/68

Gamma Radiolysis

Nitric acid solutions of varying concentrations have been gamma irradiated under a variety of conditions and have been analyzed for stable products. It was observed that the nitrite ion is the only stable major product in nitric acid solutions above 0.1 M; hydrogen peroxide is produced at a smaller rate and is consumed by reaction with nitrite. Gaseous product yields were also determined; they are extremely low except for oxygen at the higher nitric acid concentrations.

Effect of Oxygen

Nitric acid solutions from which the air (oxygen) was removed using a freeze-thaw technique in a vacuum line, were irradiated. The results at the lower acid concentrations are of interest because the nitrite yields are lower than the usual reaction mechanisms would predict. The nitrite yields without oxygen were lower than in the solutions containing oxygen. An explanation for this anomaly is being sought.

Effect of Temperature

One molar nitric acid solutions were irradiated at 80°C in a Gammacell. The apparent nitrite yields were somewhat scattered and non-linear with dose. The results, however, indicate there is very little difference, if any, between the yields at 80°C and at room temperature.

Effect of Acidity

In order to explain and verify the decrease in nitrite yields at the higher nitric acid concentrations, solutions of varying acidity and constant nitrate concentration were irradiated. The results show that the acidity is responsible for the decrease in nitrite yields. Explanations for this effect of acidity are now being sought.

Gaseous Yields

Nitric acid solutions of varying concentrations were gamma irradiated in a vacuum and the gaseous products analyzed by mass spectrometry to determine the average G values of the predominant species; O_2, H_2, N_2, NO, and total gas.

These yields and trends are in general agreement with previous results obtained by alpha radiolysis and with the simple mechanistic scheme used to describe the radiation chemistry of nitrate solutions. Oxygen and hydrogen are the primary products and even these yields are very small compared to those of the nitrite ion or hydrogen peroxide. An exception is the amount of oxygen formed at the highest acid concentration.

Alpha Radiolysis

The 60 grams of PuO_2-238 microspheres to be used as an alpha source have been received, and experimental data has been obtained on the cleaning and on the solubility of the microspheres in nitric acid solutions. This information is required for proper planning on the alpha radiolysis work. Microspheres prepared from Pu-239 were obtained from Oak Ridge (sol-gel) and from Mound (plasma tourch).

The microspheres were cleaned in nitric acid to a constant, low level of activity in the solution. Using the cleaned microspheres, solubilities were measured and found to be much less for the plasma tourch prepared material than for the sol-gel material.

5. Research Contract No. AT(10-1)-1230 $ 12,000 in FY-68

Title

Fuel Sample Stability

Institution: Idaho Nuclear Corporation

Principal Scientific Investigator: G. A. Huff

Period of Contract: 5/2/68 to 6/30/68

Background & Principle

Enriched uranium is recovered from nuclear fuels at the Idaho Chemical Processing Plant by a liquid-liquid solvent extraction system. Various analyses are obtained on the dissolved "feed" or input measurement sample prior to the extraction process. The analyses requested on the input measurement samples are dependent on the type of fuel processed. All fuels, however, are analyzed for uranium, acid and specific gravity. The most important analysis is uranium which is used in criticality and accountability control. Frequently, input measurement samples are sealed in glass vials and stored for reference because verification of the uranium concentration may be required at a later date.

The integrity of stored input measurement samples was investigated by analyzing them for the constituents previously determined. The samples used for this work had been stored for 11 to 26 months. A statistical analysis of the data showed conclusively that the uranium concentration in the stored input samples tends to increase with time. The mechanism by which this is being accomplished is yet to be defined; from an examination of the storage vial it would appear the technique of sealing and quality of the seal makes evaporation a prime suspect. However, the possibilities of precipitation, absorption and plating must also be investigated.

The existence of undissolved uranium in the sample which goes into solution during storage must also be considered; however, in view of extenuating information such as plant material balance data, it would appear that the probability of this mechanism operating to the extent of approximately a 2% increase in concentration, as reflected in the samples, is extremely remote. However, the significance of the consequences of this possibility makes it important that additional studies be made.

Specimens of dissolver solutions were obtained from reprocessing batches of representative irradiated fuel and sealed in glass vials. These samples were apportioned for periodic analysis over a period of several years, and the first analyses were made in August 1966. Results of subsequent analyses in August 1967 indicated extensive deterioration of the samples, and the experimenters decided that future analysis would be meaningless.

Objective

Develop equipment and techniques for assuring complete sealing of sample containers to eliminate any changes in dissolver samples due to evaporation.

Analyze samples of dissolved irradiated reactor fuel which have been in storage for short periods of time, i.e., two or three months, to determine if any deterioration has occurred.

Procedure

Input measurement samples from two batches of Japanese MTR type fuel processed during September are available for study. They have been stored for about 2½ months. A visual inspection shows them to contain a small amount of solids. Examination of these samples will provide an opportunity to study the solids, reduce the time variable, and check the integrity of the glass seal.

These samples will be compared to the aged samples already remeasured to aid in the determination of useful shelf life of samples.

Techniques and equipment will be developed that will permit the transferring of input samples to glass vials and the sealing of the vials under remote conditions that will insure a satisfactory seal.

Status as of 6/30/68

Remote Sealing of Glass Vials

As previously stated, the dissolved fuel input measurement samples are stored in glass vials at the Idaho Chemical Processing Plant. These vials are filled and flame-sealed with a hand-operated oxygen-propane torch behind a lead barricade. Under this condition, significant numbers of faulty seals have been obtained due to the limited working time set by AEC exposure standards and by the difficulty of the operation.

A working model of a remotely operated apparatus was designed and built for test installation in a shielded cave in the Remote Analytical Facility. The apparatus, essentially a vertically mounted lathe, consists of two synchronously rotating chucks, one holding the vial at the bottom and the other at the top of the neck. The neck is melted with an oxygen-hydrogen flame in methanol-saturated air operating at 3300°F, then the seal is formed by raising the top chuck with a remotely actuated motor. The oxygen and hydrogen are produced with a Wenes Water Welder by water electrolysis. The major task remaining is the design and fabrication of equipment to prevent the buildup and consequent explosion of unburned gases.

Analysis of Stored Samples

A set of previously stored irradiated fuel samples was analyzed for uranium concentration, specific gravity, and acidity to compare the effects of longer storage time on the stability of these samples. Evaluation of the data is in progress and the results will be reported at a later date.

6. Evaluation of Computers for Nuclear Materials Control (10/1/59)
WCAP-6016

A. R. Fritsch, H. Ginsburg

A medium-sized digital computer, with auxiliary data-logging equipment, is recommended for use in the Nuclear Materials Control System (NMCS). For the assumed nuclear complex, a manpower reduction of about eight men may be achieved at a capital cost of about \$580,000 and an additional annual operating expense of \$140,000. The increased costs can be justified by the expected improvements in NMCS operating efficiency and the greater availability of information.

Data from reactors or chemical processing plants would be recorded by conventional automatic data-logging methods. Processing of the data by the computer would include the verification of the numerous recorded readings by inter-correlation techniques, as well as recalibration of the instrumentation by statistical techniques.

7. Research Contract No. AT-33-1-GEN-53 $ 20,000
(Related to Safeguards)

Title

Radioactive Waste Containers for Storage and Shipping

Institution: Mound Laboratory, Miamisburg, Ohio

Principal Scientific Investigators:

Period of Contract: 1/1/67 to 6/30/68

Background & Principle

This work is directed toward providing disposable and/or reusable shipping containers for large volume quantities of Plutonium-contaminated wastes, both dry and liquid or semi-liquid. These containers would provide for safe storage of these materials prior to as well as during shipment, since they meet the AEC Manual Chapter 0529 hypothetical accidental conditions.

Developments made during this effort may also be applied to shipment and/or safe storage of non-discardable Plutonium compounds or Plutonium in fabricated components.

8. Bilateral Certification of Reactor Design (2/28/59) WCAP-6013

A. R. Fritsch, H. D. Hickman

The safeguards and controls clause in the Bilateral Agreements for Cooperation for the Civil Uses of Atomic Energy provides for the review of the design of any reactor made available for the cooperating nation. This report presents a number of forms suitable for the Bilateral Certification of designs for power, test, and research reactors. A set of design requirements has also been detailed.

I. Plant Protective and Monitoring Systems and Devices

Page

1. Tamper-Resistant Instrumentation System (ACDA) 207
2. Tamper-Resistant Cables for Remote Monitoring (ACDA) 209
3. Tamper-Indicating Safing System (USAEC) 210
4. Personnel Monitoring to Detect Diversion of Fissionable Materials in Chemical Processing Plants (USAEC) 211
5. Closed Circuit Television for Security and Surveillance (USAEC) 212
6. Devices and Techniques for Perimeter Protection (USAEC) 213
7. Devices and Techniques for Protection of Enclosed Spaces (USAEC) 214
8. Devices and Techniques for Access Control and Protection (USAEC) 215
9. Devices and Techniques for Protection of Pipe and Conduit (USAEC) 216
10. Alarm Transmitting Systems and the Protection of Communications (USAEC) 217
11. Miscellaneous Devices for Plant Security (USAEC) 218
12. Miscellaneous Devices for Safeguarding a Fuels Reprocessing Plant (USAEC) 219
13. General Activity Detectors for Safeguards Use (USAEC) 220
14. A Doorway Monitor for Safeguards Use (USAEC) 221

1. Research Contract No. ACDA/ST/RA-52	$ 65,000 FY68	$160,000 FY69

Title

Tamper-Resistant Instrumentation System

Institution: Sandia Corporation

Principal Scientific Investigator: V. Engle

Period of Contract: 7/1/67 to 6/30/69

Background & Principle

The Arms Control and Disarmament Agency has, in the past, been concerned with the development and application of sensors to a variety of possible arms control problems. In addition, the Agency has recently undertaken the development of a tamper-resistant data link for the secure transmission of data from a sensor to a central data recording unit. However, the Agency has not had an immediate requirement for a tamper-indicating/tamper-resistant instrumentation system that could operate unattended for relatively long periods of time in a "hostile" environment. This situation has changed with the tabling of the Non-Proliferation Treaty and the inclusion of provisions for international safeguards inspection under Article III. The requirement for a highly credible international safeguards inspection system and the objections of the most advanced non-nuclear nations to possibility of industrial espionage under the guise of safeguards inspection have combined to underscore the urgent need for unobtrusive instrumentation which can record without any alteration all of the necessary safeguards data. Quite apart from the unobtrusive features of such safeguards instrumentation, the use of this type of equipment should reduce both number of inspectors and the total cost of international inspection under the IAEA.

Objective

To develop for use in the verification of arms control agreements and the inspection of nuclear facilities placed under international safeguards an unattended, tamper-indicating/tamper-resistant instrumentation system. The system would include the secure volume technology and devices required to protect both sensors and the data recording units as well as the technology required to ensure the unaltered and uninterrupted transmission of all of relevant safeguards data to the recording unit.

Procedure

Part I

Study the problem of developing a tamper-indicating/tamper-resisting safeguards instrumentation system to be applied to declared nuclear facilities under international inspection by such organizations as the IAEA and Euratom. The study should:

a. Describe the system functionally.

b. List constituent elements and specify functional and environmental requirements.

c. Cite existing technologies and identify areas requiring additional development.

d. Identify system limitations.

e. Indicate level of effort and cost of reducing model system to practice.

Part II

Apply the design concepts developed in Part I to a specific nuclear facility, preferably under international safeguards. The selection of the facility would be based primarily on technical considerations and the complexity of the facilities.

This study will include:

a. Evaluation of the specific sensors required to safeguard the facility, the techniques for protecting the sensors and counter-measures that might be employed.

b. Design of secure data transmission techniques compatible with the rate of data acquisition required to safeguard the specific facility, drawing on previous research effort in this area.

c. Design of the secure data recording unit compatible with the data storage required to safeguard the specific facility.

d. Development, as necessary, of the necessary additional secure devices required to give the system a high degree of credibility.

e. Preparation of manpower and cost efforts required to fabricate complete system.

Part III

Fabricate, and test the tamper-indicating/tamper-resisting safeguards instrumentation system developed under Part II.

Status as of August 1968

Part I

Report completed.

Part II

Design work in progress

2. Research Contract No. ACDA/WEC/FO/RA-51 (NBS) $ 22,000 $ 68,000
ACDA/WEC/155 (BD&M) FY-68 FY-69

Title

Tamper-Resistant Cables for Remote Monitoring

Institution: Braddock, Dunn and McDonald, Inc. (BD&M) El Paso, Texas
National Bureau of Standards (NBS) Washington, D. C.

Principal Scientific Investigators: P. F. Boulay (BD&M)
F. Montgomery (NBS)

Period of Contract: 7/1/67 to 6/30/69

Background & Principle

In September 1966, BD&M completed a research investigation leading to the design of specific sensing devices capable of tamper-resistant operation while unattended. The study developed design features and principles most likely to afford at least the same operational reliability as if the sensing devices were attended by human observers or operators.

As a follow-on to the above work, BD&M is conducting an experimental study of methods of assuring the integrity of the cables to sensors located remotely from a secure data recording chamber and the sensors themselves. The use of random anomalies in cables and the reciprocal properties of some sensors will be examined as potential contributors to the problem.

Objective

Field Test the tamper-resistant cabling developed under the above mentioned contracts and determine if it can successfully resist penetration. The tests will include the actual monitoring of some aspects of a nuclear facility such as a power reactor or fuel processing plant.

Status as of August 1968

The field test is in progress and is expected to be completed in FY69.

3. Research Contract No. AT(45-1)1350 $ 135,000

Title

Tamper-Indicating Safing System

Institution: General Electric Company (HAPO)

Principal Scientific Investigators:

Period of Contract: 7/1/64 to 7/12/65

Background & Principle

The scope is indicated by the following introduction from the report:

> "It may be helpful to a monitoring team, charged with assuring itself that a given power reactor has not been used for weapons production, to know that certain components have not been manipulated between monitoring visits. This report describes a "safing" wire and a "safing" seal, previously developed at Hanford, and the manner in which they and supporting equipment may be prepared and utilized for detection of tampering."

On November 16, 1966, a demonstration of the installation of a seal on the "F" Reactor at Hanford was presented to 53 representatives from 45 nations and international organizations.

Work continued in 1967 to apply the safing wires and seals to a shutdown reactor as a field test of their effectiveness.

4. Research Contract No. $ 20,000

Title

Personnel Monitoring To Detect Diversion of Fissionable Materials in Chemical Processing Plants

Institution: Argonne National Laboratory

Principal Scientific Investigators: D. Engelkemeir, A. H. Jaffey

Period of Contract: 1956-1957

Results

This study resulted in the preparation of a report entitled "On the Possibility of Personnel Monitoring to Detect Diversion of Fissionable Materials in Chemical Processing Plants", May 1957. Consideration was given only to methods for detection of materials carried by individuals. The study proposes to load fissionable materials with gamma-emitting or neutron-emitting substances which may be detected by a group of energy-discriminating sodium iodide detectors or by a battery of BF_3 counters. Isotopic tracers U-232 and Pu-240 are proposed. About 100 ppm of U-232 would be used to spike uranium and the 2.6 Mev gamma-emitted would be measured. Plutonium would be detected by measuring spontaneous fission neutrons from Pu-240 added to the extent of about 5% by weight. The report states that such "spikes" would permit the detection of 10 grams of uranium or plutonium.

The major design features of a personnel "walk through" detection facility, estimated to cost $77,000 exclusive of the building, are outlined.

5. Closed Circuit Television for Security and Surveillance (10/1/59)

WCAP-6017

G. Kivenson

Television has been studied as a means of industrial surveillance. The practicality of the method has been established, and certain optimum operating conditions have been determined.

An alarm utilizing closed circuit television has been developed and tested.

Evaluations were made of the Dage fixed camera system and the General Precision Laboratory remote control camera. Maintenance requirements for the equipment were moderate during the eight months of testing. Improvements in the vidicon camera tubes or replacement by image orthicons would appear desirable to ensure good performance.

The General Precision camera, using a 600 line scan, was capable of higher resolution than than the Dage system which employs 400 lines. For identification work, it is felt that the additional cost of the 600 line scan is justified.

6. Devices and Techniques for Perimeter Protection (10/1/59)
WCAP-6018

J. N. Ellyson

The various types of penetrations against which perimeters must be protected are examined in this report. The techniques or systems which were considered for perimeter security applications are described briefly; included are: low frequency radio energy, UHF and microwave equipment, photoelectric systems, sound beams, and closed circuit television.

Experiments were conducted with three types of perimeter protection equipment: a photoelectric system, a low frequency electromagnetic system, and a sound beam apparatus. The equipment used and the results obtained are described.

It was concluded that photoelectric systems of current design are generally inadequate for perimeter security work. The low frequency radio system was found to be satisfactory except for some difficulty with spurious alarms and variable sensitivity.

Comparatively little equipment intended primarily for perimeter security is commercially available. Therefore, further investigation of a number of promising techniques is recommended.

7. Devices and Techniques for Protection of Enclosed Spaces (10/1/59)
WCAP-6019

G. Kivenson, J. N. Ellyson

Methods for the protection of enclosed spaces such as rooms, vaults, and passageways have been studied and evaluated.

Radio, ultrasonic, and light beam devices were investigated. The ultrasonic units appeared to give the maximum protection in terms of coverage, although initial cost is high.

Very little work has been done in the past on the development of standardized testing methods for this type of apparatus. Comparison is made, therefore, on the basis of range, cost, and type of service for which the apparatus is suitable. A table listing the equipment studied and some of its properties is included.

A number of promising possibilities for the improvement of enclosed space protection apparatus have arisen during the present work. These include the use of code modulated radio systems, far infra-red devices, and passive photomultiplier units. Further evaluation would be desirable.

8. Devices and Techniques for Access Control and Protection

(10/1/59) WCAP-6020

G. Kivenson

The problem of protecting gates, doorways, and window opernings in plant sites operating under a Nuclear Materials Control System (NMCS) is discussed from two standpoints: automatic alarms for indicating unauthorized entry, and gate pass systems for maintaining overall security with a minimum guard force. This report assumes that normal locking procedures can be readily defeated.

Qualitative results obtained with door and window alarms of various kinds are described. Vibration detectors are an inexpensive and readily-applied method for windows. Photocell systems are relatively costly and easily defeated when used for doorways. Radio frequency methods, while not fully developed for these purposes, seem quite promising.

Several automatic gate pass systems are described. Two of these, designed during the NMCS program, appear to overcome some of the objections of commerically available pass systems.

9. Devices and Techniques for Protection of Pipe and Conduit

(10/1/59) WCAP-6021

G. Kivenson, J. N. Ellyson

The original Nuclear Materials Control System (NMCS) study indicated that it would be necessary to specify protection schemes for various installations of pipe and conduit. No suitable commercial equipment was available for this purpose. Systems for the protection of pipe and conduit were therefore designed and tested, subject to the limitations imposed by a premature termination of this portion of the NMCS Project. Four of the most promising methods of protection were selected for detailed evaluation. Each of the four was examined by means of an experimental prototype. The test results with each prototype are described.

Recommendations are made for the use of each device in typical field installations. It is concluded that the devices described can protect most types of pipe and conduit installations found in a nuclear complex.

Sonic or proximity alarms give best protection. An evacuated jacket thermocouple gauge alarm system appears practicable.

10. Alarm Transmitting Systems and the Protection of Communications

(10/1/59) WCAP-6022

G. Kivenson, J. N. Ellyson

The application of a Nuclear Materials Control System (NMCS) to a nuclear complex requires protected communications systems for the efficient and secure transfer of many types of information. The distances involved may range from a few feet to several hundred miles.

Many of the requirements can be met by techniques well known to government agencies and the military (e.g., cryptographic systems). Most alarm transmission systems are intimately connected with the operation of a specific protection device. Therefore, the work planned by the NMCS Project in the field of communication security and alarm transmission was limited to:

a. A study of techniques and apparatus for generating identification and security condition signals, in certain geographical areas;

b. A special coding system for use in security condition reporting;

c. Evaluation of a commercial system for the protection of wire transmission links;

d. Methods of speech scrambling;

e. Special short-range alarm transmission systems.

It was concluded that equipment is available to assure security of communications by radio or wire.

11. Miscellaneous Devices for Plant Security (10/1/59) WCAP-6023

G. Kivenson, J. N. Ellyson

To fill the need for auxiliary protective equipment to be used for a Nuclear Materials Control System (NMCS) in moderately sensitive areas, an inquiry was made into the availability of small, relatively, inexpensive devices having a wide range of applications. These units included vibration alarms, power line monitors, and mechanical disturbance indicators. Equipment for detecting pneumatic line failure and unauthorized valve operation was also evaluated.

Some commercially-available equipment was found to be adaptable for these purposes. It was necessary, however, to design a number of new devices for certain specific applications. Modified devices should be quite practicable.

12. Miscellaneous Devices for Safeguarding a Fuels Reprocessing Plant (10/1/59) WCAP-6024

W. E. Foster

Various anti-syphon devices, a temperature recorder, and a device to assure a constant rate of air flow in pneumatic instrument lines were evaluated for possible inclusion in a Nuclear Materials Control System (NMCS). The devices are intended for use in a fuels reprocessing plant as supplementary equipment for the prevention of unauthorized diversion of source and special (SS) material. The plant is assumed to be under stringent inventory control conditions.

Recommendations concerned with the application of the devices to a fuels reprocessing plant are presented.

13. General Activity Detectors for Safeguards Use (10/1/59)
WCAP-6029

F. J. Arsenault

The application of radioactivity monitors to the detection of SS material diversion is discussed. Slug counters on the dissolver slug chutes at chemical processing plants are currently in use and appear practical. Pipeline monitors, area monitors, and portable monitors to survey areas, personnel, and vehicles are considered. It is concluded that a self-contained detector-alarm unit could be developed for fuel slug counting, pipeline and area monitoring.

A Doorway Monitor for Safeguards Use (2/1/60) WCAP-6038

F. J. Arsenault

A Doorway Monitor has been developed as part of a Nuclear Materials Control System (NMCS) to discourage the removal of source and special (SS) nuclear materials from a plant or work area by concealment on the person. The human relations problems involved have resulted in an emphasis on passive detection.

The effort has been divided among three methods of detection:

a. detection of the spontaneous fission neutrons from plutonium-240;

b. detection of the 90 and 184 kev gamma-rays from uranium-235;

c. detection of metal shielding.

An array of BF_3 proportional counters and plastic scintillators is proposed to detect the passage of plutonium or U-235 through a doorway.

J. Process Instrumentation

Page

1. Methods for Estimating the Integrated Thermal Power Produced in a Nuclear Power Reactor (IAEA) 298/RB 225

2. Feasibility and Design Study of a Method to Record the Quantity of Fuel Charged and Discharged From Nuclear Reactors (IAEA) 299/RB 228

3. Integrated Power Monitor for Safeguards (IAEA) 576/RO 231

4. Metrology of Nuclear Materials (USAEC) 232

5. In-Line Monitor for Fissionable Material (USAEC) 235

6. Volume and Density Measurements Using Ashcroft Pressure Gages (USAEC) 237

7. Volume and Density Measurements on Radioactive Solutions (USAEC) 238

8. Recording Wattmeters for Safeguards Use (USAEC) 239

9. Neutron Flux Monitors for Safeguards Use (USAEC) 240

10. A Thermal Power Meter for Safeguards Use (USAEC) 241

11. Continuous Monitors for Uranium and Plutonium in a Chemical Processing Plant (USAEC) 242

12. Specific Gravity Instruments for a Nuclear Fuels Reprocessing Plant (USAEC) 243

13. Liquid Level Instruments for a Nuclear Fuels Reprocessing Plant (USAEC) 244

14. Flowmeters for a Nuclear Fuels Reprocessing Plant (USAEC) 245

15. Evaluation of N-16 Monitoring in WTR Primary Coolant for Measurement Flow and Reactor Power (USAEC) 246

1. Research Contract No. 298 $ 30,000

Title

Methods for Estimating the Integrated Thermal Power
Produced in a Nuclear Power Reactor

Institution: Stanford Research Institute
California, U.S.A.

Principal Scientific Investigators: E. M. Kinderman, S. Softky

Period of Contract: 12/15/64 to 3/15/66

Background & Principle

The IAEA safeguards system is designed to ensure that nuclear materials, equipment, and facilities subject to the Agency's control are used only for peaceful purposes. In inplementing the system, the Agency may require that facilities contain enough instrumentation to permit a determination of what is happening in a closed part of a reactor or processing plant. IAEA inspectors may have access at all times to all places and to data as necessary to account for special nuclear materials supplied or for fissionable materials produced.

Among the more important facilities subject to safeguards inspections are nuclear reactors designed to produce electric power. A method is needed for the inspector to estimate, independently of other measurements, the total energy released by the nuclear reactor during the period since his last inspection; this period may, in practice, range from a few months to a few years. The method ideally should be one which (1) does not interfere in any way with the operation of the reactor facility and (2) cannot be tampered with.

Procedure

A photovoltaic cell in combination with its integrator has been developed as a potential monitoring system for the integrated power generated by power reactors.

The photovoltaic cell consists of a piece of anodized tantalum, with a gold counterelectrode evaporated on the anodic insulating film. When this device is irradiated with gamma radiation, it generates a current in the same manner as an illuminated solar cell. Such a radiation monitor appears simple to use since the photovoltaic cell can be small and easily inserted deep into the shield, outside the reflector of a reactor, where gamma radiation is proportional to reactor power. The integrating device can be left in a more accessible spot for readout with wires connecting it to the detector.

The detectors are reasonably linear in current output vs gamma-radiation intensity; a commerical electrolytic type of current integrator adequately integrates current from the detectors; a method for fabricating detectors with adequate long term (over six months) stability and reliability in gamma radiation fields has been developed.

The general applicability of the photovoltaic monitor system, for period up to six months' continuous exposure, has been demonstrated. Since the monitor generates a current proportional to radiation intensity if it is placed in the radiation field in or near the reactor shield (so that radiation intensity at that point is proportional to reactor power), the monitor can be calibrated to read reactor power in terms of output current. Since it requires no power supply, it can be left unattended for long periods; a reading of integrated current will yield a measurement of integrated reactor power.

Conclusions

The results obtained in fabrication and testing of the latest photovoltaic cells, and the successful performance of the latest type of current integrator in conjunction with the monitor, show that the system performs as expected; i.e., the photovoltaic monitor and its integrator should work satisfactorily as a passive reactor power monitor system. The fluctuations at low current indicate that the monitor will be useful from 10^4 rad/hr to at least 10^6 rad/hr dose rate of gamma radiation. Life tests on this type of photocell indicate that its life expectancy is at least 3 x 10^8 -rad total dose, although it would require one-year irradiations of the cells made by latest techniques to prove that reciprocity holds with regard to the dose rate-time product.

The monitor could be used in a position deep in the biological shield of a reactor, or even inside the shield (and outside the reflector), wherever a dose rate somewhat in excess of 10^4 rad/hr of prompt gamma radiation can be obtained at expected power levels. Low leakage twin lead-shielded cable would be used from the detector to the current integrator in accessible spot outside the shield. The current integrator can be easily readout on-the-spot by a portable device. It includes a constant current source, a digital clock, and a triggering circuit actuated by the integrator when it discharged, so that the clock is stopped, yielding the (current) x (time) product remaining in the integrator.

The photovoltaic system is intended as a monitor to check the accuracy of reactor operating records. One would expect that falsification of the operating records would be accompanied by attempts to tamper with the monitor. Techniques to prevent successful tampering are discussed below.

A dummy cable attached to another current integrator could be included inside the same cable shield. The reading on the second integrator would indicate any electrical pickup or artificially induced currents in the two cables. Indicators that recorded the maximum and minimum temperatures to which the system had been exposed might also be used. These would indicate thermal tampering.

Nonreproducible seals could be used where necessary to give evidence of mechanical tampering or outright disassembly.

The unit could be most easily calibrated with a direct-reading, ultrasensitive microammeter (Greibach or equivalent), at a time when the reactor was being run at a suitable power level. This type of instrumentation is portable and rugged; it could be easily carried by hand on commerical aircraft.

Before photovoltaic monitors are incorporated in engineered systems, further measurements are required. First, the monitor detector should be used in the actual radiation field expected outside the reflector of a large reactor to ascertain whether the neutrons in the radiation significantly shorten the monitor's lifetime or affect its calibrations. Second, the monitors should be irradiated for at least a year at low dose rate (less than 3×10^4 rad/hr) in order to verify that actual (not merely total dose) lifetime is satisfactory. This lifetime test should use the E-Cell type current integrators, to provide lifetime tests of this portion of the apparatus as well.

2. Research Contract No. 299 $ 14,020

Title

Feasibility and Design of a Method to Record the Quantity of Fuel Charged and Discharged From Nuclear Reactors

Institution: United Kingdom Atomic Energy Authority

Principal Scientific Investigators: E. P. Hotchen, C. E. Brindley

Period of Contract: 1964 to 1966

Background & Principle

"The International Atomic Energy Agency's procedures for safeguarding large power reactors give their inspectors right of access to the reactor sites at all times. With the increasing proliferation of power reactors however, there is a need for a system of inspection which is effective with limited and intermittent visits to sites. Such a system should be practicable at reactor sites which are refuelled intermittently, since in principle at least, inspectors can arrange to be present during refuelling. Where reactors are continuously refuelled it may be possible to use some kind of monitoring system to give surveillance during the periods when inspecting staff are absent. Ideally, the quantity and quality of fissile material produced should be monitored, but surveys of the possibilities of making nondestructive assays of plutonium in irradiated fuel have shown that no fully adequate method of measurement is available at present. Plutonium production may however be estimated by correlation with burnup, and this in turn may be estimated from measurements of irradiation time if power distribution and history are known.

"In principle it is feasible to monitor irradiation time fairly accurately by means of attachments to, or samples from, individual fuel elements. However by reason of the large number of fuel elements in some reactor designs, the complexity of refuelling machinery and the scale of inspecting effort required to support such a procedure, it is appropriate to consider the merit of more approximate measurements made by sensing devices external to the reactor core. Since in large power reactors the distribution of rating does not vary rapidly in normal steady operation it is probably acceptable to determine irradiation time using sensing devices located at core access positions. These devices may be independent fuel movement monitors or detectors supplying information to a central processing and recording instrument. If it be objected that such a system is still too complicated and too costly then it is necessary to remove a further step from the core, and site a much smaller number of independent monitors on the fuel ingress/egress facilities and also the reactor refuelling machinery. The machinery locations offer a check between core refuelling quantity and facility utilization. This simplified system does not retain the connotation with core location so that this must be monitored additionally using perhaps signals from the refuelling machinery or photographic methods. This expedient would be vulnerable to tampering but in the longer term the trend to automated refuelling may allow reliable monitoring based on secure facsimilies of refuelling instructions.

"Since the difference between systems using monitoring instruments at core access locations or refuelling area access locations is principally one of degree, attention was directed to the requirement for a monitoring instrument which can reliably detect fuel movement."

Procedure

A survey was made of available gamma radiation sensing devices, and a prototype refuelling monitor was designed, constructed and tested. The scope of the work is indicated by the table of contents of the report:

- Detection of Fuel Movement

- Neutron Detection

- Gamma Radiation Detection

    - Photon Detection
    - Gas Ionization Devices
    - Photocells and Photomultipliers
    - Semiconductors
    - Appraisal of Gamma Radiation Detectors

- Refuelling Monitor Design

    - Logical Performance
    - Information Recording
    - Discrimination Against Nonfuel
    - Detection Threshold Setting
    - Reliability

- Application of Monitoring to Reactor Designs

    - The Access Point System
    - The Refuelling Machine System

- Design and Testing of a Prototype

    - Description
    - Performance

In order to test the feasibility of a basic refuelling rate monitoring instrument, a prototype was constructed to demonstrate the effectiveness of the proposed detector, the adequacy of the logic circuitry and to explore the effects of sensor spacing and speed of fuel element travel.

Conclusions

"It has been argued a refuelling rate monitoring system applied to a power reactor constitutes a useful physical check with which to supplement an audit of fissile material production. The indirectness of its relevance to estimation of plutonium production and the need for supporting information on flux distribution, power history, etc. is accepted."

Two basic assumptions have been introduced: first that the monitoring system should function in a hostile environment, and second, that it should have an unattended lifetime of six months. A specification for a monitoring system was prepared and a survey of the means available for meeting the requirements of the specification has led to several basic general conclusions.

One of the several conclusions, the following appears to be of primary importance:

> "Of the available bases for sensing the presence of reactor fuel elements that which depends on the gamma radiation from fission products is preferred. The chief reasons for this are
>
> a. the unequivocal nature of this characteristic of irradiated fuel;
>
> b. the difficulty of simulating such a signal artifically;
>
> c. the relatively wide range of suitable detection devices;
>
> d. the large margin on detector sensitivity available by reason of the high signal intensity.
>
> It follows that the concept of refuelling monitoring has had to be restricted to counting the movement of irradiated fuel. It has been argued that if this is done, the movement of new fuel can be consequently inferred. An advantage of monitoring irradiated fuel is that special handling precautions are necessary which cause the movement of the material to be restricted to prepared channels."

The response and logic action of a prototype refuelling rate monitoring instrument was satisfactorily demonstrated using an artificial gamma source. The unit reacted correctly to all of the accept and reject sequences.

3. Research Contract No. 567 $ 5,000

Title

Integrated Power Monitor for Safeguards

Institution: South African Atomic Energy Board's National Nuclear Research Centre at Pelindaba, Transvaal, South Africa

Principal Scientific Investigator: W. H. du Preez

Period of Contract: 11/1/67 to 11/1/68

Background & Principle

The evaluation of the power output of a water cooled reactor is most surely done by absolute means - the product of coolant flow rate and Δ T. However, this is at present done manually and at fixed intervals of time. The refinement of flow and ΔT measurements is being done at SAFARI I to improve this calculation. The safeguards point of view is that no records are maintained which are independent of the plant operators.

Procedure

The programme of work shall be the design and construction of an instrument which, when installed in the primary coolant circuit of a water-cooled reactor, accepts one of two standard signals (1-5 MA, or 0-10V) for both the Δ T and the coolant flow rate, and integrates the values received on a continuous basis to indicate a quantity of energy produced by digital means. Provisions will be provided so that the instrument will operate independently during normal power failure for a period of at least four months. The electronics providing the multiplication, integration and print-out functions will be an independent and sealed unit.

The programme is expected to be completed in one year. The total cost of the project is estimated as $10,000, of which $5,000 (50%) is required from the Agency. The rest will be borne by South Africa Atomic Energy National Nuclear Research Centre at Pelindaba.

4. Research Contract No. NBS Project No. 2120430 $ 10,000 to 6/30/68

Title

Metrology of Nuclear Materials

Institution: National Bureau of Standards

Principal Scientific Investigator: P. Pontius

Period of Contract: 1/1/68 to (continuing)

Background & Principle

The Metrology Division of the National Bureau of Standards has extensive experience in the field of measurement techniques and the statistical control of measurements.

Measurements, particularly weights, are of prime importance in the inventory control of special nuclear materials. It is possible that modern measurement techniques may be applied to safeguards procedures to improve the accuracy of inventory control. Accordingly, the Office of Safeguards and Materials Management (AEC) has awarded a contract to the National Bureau of Standards to undertake a study to determine what improvements may be made in measurement procedures used for safeguards.

Objective

This study will be addressed to two aspects of measurements for safeguards:

1. The application of modern measurement techniques and statistical control of measurement such as have been developed and applied in other fields. This investigation will lead to specific recommendations for improving the measurement techniques now in use or to the conclusion that they cannot be appreciably improved without great expense.
2. Evaluate novel methods of measurement which are presently under development or which may have to be specially developed for this study. This aspect of the study will be exploratory in nature and may, or may not, result in new and novel techniques of value to safeguards procedures.

Procedure

The Metrology Division

(1) will review in depth one area in which accountability depends on extrapolating the results of analysis to large volumes of liquid, such as in the leaching process at the head end of a spent fuel processing plant. This review will include the entire procedure (i.e., calibration of

tank, instrumentation, sampling, analytical tests, etc.) with the location of margin sources of variability in the accountability figures as the objective. Data analysis will be done on such collections of suitable data as can be located. Various NBS staff members from other disciplines will be requested to assist in detail analysis as appropriate.

(2) will review in depth one or more output-input steps in which accountability is determined exclusively by weighing techniques. This review will include the manner in which requirements have been established, the process, and the evaluation of results.

(3) will review one or more areas in which weighing is one step in the overall accountability, such as the weighing of pellets, rods, scrap, etc. This review will include the manner in which requirements have been established, the process, and the evaluation of results.

(4) will also investigate the application of holographic and other techniques to the security of sealed or fabricated items, etc., such as may be submitted by the AEC for study and the application of optical techniques to the determination of such characteristics of material as density of fluid, etc.

Status as of 10/1/68

A trip was made to the West Valley, N. Y. chemical processing plant of Nuclear Fuel Services, Inc., to obtain process information to supplement that obtained during earlier visits to Richland, Washington, and to Idaho Falls, Idaho. Based on the observations made, the following areas and activities are being examined to identify possible improvements:

1. Accountability Figures

Beginning and ending material inventory figures are developed from sampling numerous inventory tanks other than the input accountability tank. These tanks may or may not be instrumented in the same manner as the accountability tank.

Since the process solutions are very dilute, there is some question regarding the effect of extrapolation of the uncertainty of the analytical procedures. The same type of problem also exists in the measurement of waste solutions and requires examination.

2. Consistency of Inter-Plant Measurements

The consistency of material measurements between plants (shipper/receiver data) requires a standardization not only of methods but also a standardization of detailed procedures. It is possible to monitor a precise process with another less precise process, but the less precise figures should not be used for accountability. Provided that realistic uncertainties are available for both measurement processes, such a method of monitoring may offer a significant economical advantage for safeguards.

3. Process Tank Calibration

   Present methods of calibrating process tanks appear to be very time consuming, and, further, it is almost impossible to verify the results after a plant has been in operation. The use of transfer standards, such as volumetric provers, would not only shorten the initial calibration time, but would also permit periodic recalibration. These techniques have been used widely in the petroleum industry, and could be applied for safeguards purposes to chemical processing plants.

4. Automated Measurement Processes

   It appears that some attention should be directed toward automating measurement processes. Automated measurement processes not only assure consistency but also tend to reduce manpower requirements.

5. Holographic Techniques

   It has been established that holographic techniques at the present time have no advantage over other established techniques for identifying fuel elements. Existing techniques are not applicable to large objects nor to objects which are apt to distort in excess of a wavelength of light in normal operation.

   Holography may be useful in the identification of small objects (such as seals) and does provide a means whereby a single negative can be subdivided without significant loss of resolution. Parts of the same negative, for example, could be sent to different locations for use in object identification. Such techniques are well established and need no further development for safeguard uses.

5. Research Contract No. AT(10-1)-1230 $ 3,000 in FY-68

Title

In-Line Monitor for Fissionable Material

Institution: Idaho Nuclear Corporation

Principal Scientific Investigators: G. L. Booman, J. E. Rein

Period of Contract: 5/2/68 to 6/30/68

Background & Principle

The safeguards control of fissionable uranium and plutonium material in a chemical processing plant is best attained by continuous monitoring of key process points such as the line from the dissolver to the input tank, waste streams, and the final product streams. Additional benefits are improved plant efficiency for the overall recovery of fissionable material by providing the warning of process malfunctions and by preventing loss of material caused by above-critical levels flowing to a vessel which is not criticality safe.

Objective

Develop a tamperproof in-line monitor to automatically transmit chemical reprocessing plant process data to a computer. For this feasibility study only: design flow rate and tank volume measuring system for operating model. Define interfacing with computer, and optimize shielding requirement.

Procedure

1. Develop shielding requirements
2. Design irradiation and measuring chambers.
3. Develop methods for simultaneous measurement of uranium and plutonium.
4. Develop and test on-line digital computer data acquisition, reduction, and display system.
5. Install operational model of fissionable isotope monitor.

Status as of 6/30/68

This limited feasibility study was to determine if the proposed monitoring system merits further effort and, if so, to outline the steps of a developmental program.

A laboratory mockup of a monitor system has been developed and successfully tested. The basis of operation is irradiation of the flowing stream with neutrons from a Pu-238 beryllium source in one chamber, then measurement of the fission product delayed neutrons in a second chamber. A small, on-line digital computer provides immediate information output of fissionable isotope concentration levels, trend predictions, integrated fissionable isotope contents in the receiving vessel, criticality alarm, and has self-checking functions for tamperproof operation. The last can include control to detect the operation of flow diversion devices such as a bypass valve system. The mockup model includes the means to monitor the neutron flux level at the surface of the irradiation chamber which provides a correction for neutron absorbing components in the sample stream.

Shielding

Work was begun on the design of shielding configurations which could be used to define optimum earth, concrete, and water shields. The first experiments could be made with earth shields by placing a single boron trifluoride detector tube at various distances from a 238-Pu-Be neutron source having an emission rate of about 3×10^8 neutrons/sec. Holes could be made in the ground outside the chemical processing plant for this experiment.

Data Acquisition

Design of interface circuits to permit input of stream flow and holding tank volume information to the on-line computer was completed and drawings were prepared. These additional inputs will improve the accuracy of fissionable material measurement and will permit criticality margin calculations as part of the computer program.

6. Research Contract No. AT-33-1-GEN-53 (Related to Safeguards) $ 5,000

Title

Volume and Density Measurements Using Ashcroft Pressure Gages

Institution: Mound Laboratory, Miamisburg, Ohio

Principal Scientific Investigators: D. F. Luthy, R. L. Fiely

Period of Contract: 1/1/67 to 6/30/68

Results

An experimental tank of lucite was used so actual volume could be measured. The tank contained two dip lines, one to the bottom and another a known distance from the bottom of the tank. By using the Ashcroft gage in the standard way and making the change in pressure from the two dip lines, a density for the solution could be calculated. This density was then used to correct the standard Ashcroft gage reading. Percent error by this method, over the range studied, ranged from 1.95 to 5.54 in comparison to the standard readings which ranged from 3.71 to 30.0 percent.

7. Research Contract No. AT-33-1-GEN-53 (Related to Safeguards) $ 5,000

Title

Volume and Density Measurements on Radioactive Solutions

Institution: Mound Laboratory, Miamisburg, Ohio

Principal Scientific Investigators: D. F. Luthy, F. E. Morgan

Period of Contract: 6/30/66 to 6/30/68

Results

Volume of nitric acid solution containing radio-isotopes are found by using a Dynatrol continuous monitor for density and recorded on a graph where the average density can be determined in various ways. This same solution is then weighed in a suspended tank in a glovebox which is separated from the exterior of the box by a 0.025 inch Viton film. The platform balance is placed under the tank and corrected volumes of the solution weighed can be calculated. A load cell can also be used in place of a platform balance.

8. Recording Wattmeters For Safeguards Use (10/1/59) WCAP-6026

R. E. Kronk

Commercially available wattmeters can be made tamper-resistant for monitoring the electrical power produced by a nuclear power station.

9. Neutron Flux Monitors for Safeguards Use (10/1/59) WCAP-6027

R. E. Kronk

The overall objective of the Nuclear Studies portion of the Nuclear Materials Control System (NMCS) program is to predict, by computation and measurement, the fissile material inventory at a nuclear reactor site. This report is concerned with the design of NMCS neutron monitoring instrumentation, and with some of the related technical difficulties in the application of these devices to fissile material control. Two instrument systems are described which supply potentiometer recorded and digital printed outputs corresponding to the monitored neutron flux. The fissile material content of the reactor can then be computed.

10. A Thermal Power Meter for Safeguards Use (10/1/59) WCAP-6028

C. C. Webster

The proposed Nuclear Materials Control System (NMCS) requires an accurate knowledge of the net quantities of fissile materials discharged from a reactor. A thermal power meter located in the primary coolant lines of both power reactors and large test reactors is the most reliable method currently available for measuring the thermal power output of such reactors. This proposed design consists of a flow measuring device, combined with platinum resistance thermometers for measuring the temperature difference across the reactor or its associated steam generator. Tamper-resistant features are specified in order to assure the integrity of the measurements. Fissile material content of the reactor can be calculated from the data obtained.

11. Continuous Monitors for Uranium and Plutonium in a Chemical Processing Plant (10/1/59) WCAP-6031

E. R. Rosal, W. E. Foster

Various instruments suitable for the continuous monitoring of plutonium and uranium in a nuclear fuels reprocessing plant under the jurisdiction of a Nuclear Materials Control System (NMCS) were evaluated. The program emphasized the selection of plant-proven instruments which would provide independent and reliable supplementary information concerning the plant inventory. Descriptions and specifications for each instrument and its recommended points of application in the reprocessing plant are given. Tamper-resistant features to protect the integrity of the instrument readings are discussed.

The instruments evaluated include polarographs, alpha monitors, flow colorimeters, gamma-ray absorptometers and neutron monitors.

12. Specific Gravity Instruments for a Nuclear Fuels Reprocessing Plant

(12/1/59) WCAP-6033

W. E. Foster, E. R. Rosal

Specific gravity instrumentation suitable for a nuclear fuels reprocessing plant was evaluated for possible use in a Nuclear Materials Control System. Two basic types of instruments were considered:

a. pneumatic "bubbler" instruments intended primarily for use in process tanks;

b. pipe-line units for continuous monitoring of process streams.

Pneumatic instrument systems using both a two and a four dip-tube assembly were built and installed in the feed and product tank respectively of the NMCS Test Loop. The two tube system was tested on aqueous solutions of uranyl nitrate in nitric acid. At prime accountability points of a chemical processing plant under stringent inventory control, two assemblies with four dip-tubes, dual sensing units, and appropriate tamper-resistant features are recommended.

A pipe-line instrument for monitoring the specific gravity of process streams on a continuous basis ("Densitrol") was purchased and installed on the NMCS Test Loop. The instrument appears promising for further evaluation as a continuous monitor of process stream density, or as a supplement to an in-line analytical instrument. Appropriate tamper-resistant features are specified for use with the Densitrol when it is included in a Nuclear Materials Control System.

13. Liquid Level Instruments for a Nuclear Fuels Reprocessing Plant
(12/1/59) WCAP-6034

E. R. Rosal

Liquid level instrumentation for a nuclear fuels reprocessing plant was evaluated for possible application in a Nuclear Materials Control System (NMCS). Two basic types of instruments were considered:

a. pneumatic "bubbler" systems, intended primarily for use with radioactive solutions;

b. Fielden (capacitance) probes, intended for use with decontaminated product solutions.

A pneumatic instrument system using a two dip-tube assembly was built and installed in the product tank of the NMCS Test Loop and tested with various aqueous solutions of uranyl nitrate in nitric acid. Methods for calibration of tank volumes and liquid level instruments are discussed. A four dip-tube assembly with dual sensing elements and appropriate tamper-resistant features is recommended for NMCS use.

The Fielden capacitance instrument has been extensively tested at the Savannah River Plant where it was found to be vulnerable to radiation damage. Therefore, it can be used only with decontaminated material. Shielded capacitance probes with appropriate tamper-resistant features are recommended for NMCS use.

14. Flowmeters for a Nuclear Fuels Reprocessing Plant (12/1/59)
WCAP-6035

E. R. Rosal

Flow measurement instrumentation for use in a nuclear fuels reprocessing plant was evaluated for possible application in a Nuclear Materials Control System (NMCS). Two types of flowmeters were considered:

a. variable area flowmeters, primarily intended for use with radioactive solutions;

b. magnetic flowmeters

Both types of instruments have been used in several AEC reprocessing plants. The rotameter has seen service in both radioactive and nonradioactive streams, while the magnetic flowmeter has been used in decontaminated product streams.

The meters were tested under both pulsating and continuous flow conditions. It was found that these instruments are capable of monitoring continuous flow streams, but cannot give an accurate flow measurement under extreme pulsating conditions.

Leaks around the electrodes are a major problem in small flowmeters of the magnetic type. Further work should be done to increase the radiation-resistance of magnetic flowmeters, and efforts should be made to correct the leaks around the electrodes. Nevertheless, a magnetic flowmeter can be used to measure flow rates, under both moderately pulsating or continuous flow conditions, with a precision of better than 1.5 percent of full scale.

Recommendations are made for tamper-resisting both types of instruments.

15. Evaluation of N-16 Monitoring in WTR Primary Coolant for Measurement of Flow and Reactor Power (2/28/61) WCAP-6045

R. E. Kronk

The feasibility of measuring reactor power and coolant flow rate by monitoring the nitrogen-16 activity in the coolant water has been established through experiments at the Westinghouse Testing Reactor. When compared to conventional instruments for measuring power and flow rate, the coolant activity monitors show deviations of approximately ± 15%. Extensive comparisons are made for flow rates from 7,000 to 12,350 gallons per minute. Possible reasons for the deviations are discussed. Self-checking features of primary coolant monitoring instruments are also discussed and compared with available data.

A limited number of measurements made on the nitrogen-17 coolant activities show results similar to those obtained with N-16 radiation monitors.

K. Reactivity Measurements — Page

1. Preliminary Considerations in the Design of the Westinghouse Variable Loading Test Reactor (USAEC) 249
2. The Feasibility of an Annular Core Reactor as a Reactor Fuels Measuring Device (USAEC) 250
3. The Feasibility of an Annular Nuclear Core as a Reactor Fuels Measuring Facility (USAEC) 251
4. A Design for the Evaluation of a Reactor Fuels Measurement Facility (USAEC) 252
5. Experimental Confirmation of the Feasibility of the Variable Loading Test Reactor Concept (USAEC) 253
6. Assay Capabilities of a Reactor Fuels Measuring Facility (USAEC) 254
7. Decay Scheme Studies (USAEC) 255
8. Fissile Material Content by Reactivity Change-Unirradiated Fuel (BMwF) 257

1. Preliminary Considerations in the Design of the Westinghouse Variable Loading Test Reactor (4/15/59) WCAP-1161

J. A. Vreeland

A review of perturbation theory is presented specifically directed toward conclusions relevant to the design of a Variable Loading Test Reactor. This reactor is intended to produce reactivity measurements which can detect variations in fissile content of power reactor fuel elements, both with and without burnable poisons, by comparison with a standard.

The perturbation theory leads to the conclusion that the effects on reactivity of the absorption of poison and of fuel in the same spectral region can be distinguished. The extent to which these two effects can be separated is dependent upon the measurement of the reactivity effects under two conditions where the ratio of the importance function of the absorption energy to the importance function of the energies of the fission spectrum is substantially different.

The equations defining the importance functions are then used to note explicitly the dependence of these functions on the various reactor parameters. From this dependence is then inferred variations in these parameters which might change the functions enough to produce the separation we seek. Such inferences are made for both "small" and "large" assemblies where small and large are distinguished by whether changes exterior to the assembly affect the flux and importance distribution at the center of the assembly.

2. The Feasibility of an Annular Core Reactor as a Reactor Fuels Measuring Device (5/31/59) WCAP-1192

G. E. Putnam

The possibilities of determining changes in the U-235 content of large fuel elements such as that of Yankee were investigated with respect to using an annular core reactor to perform reactivity measurements which compare a given fuel element to a standard element of known composition. The sources of error are analyzed and reported.

241

3. The Feasibility of an Annular Nuclear Core as a Reactor Fuels Measuring Facility (9/8/59) WCAP-1221

J. A. Vreeland

The sensitivity to changes in fuel content of a large, low enrichment fuel assembly is studied for two measurements. These two measurements are, respectively, moderated with water and with a saturated solution of boric acid. It is shown that to conceal the removal of a fraction of the U-235, it is necessary to remove 5.5 times that fraction of the U-238. It is similarly shown that to conceal the addition of a fraction of U-235, it is necessary to replace one half of that fraction of the U-238 with gold or some comparable material.

4. A Design for the Evaluation of a Reactor Fuels Measurement Facility (6/24/59) WCAP-1232

W. J. Rusnack

The purpose of the Reactor Fuels Measurement Facility (RFMF) is to make nondestructive measurements of fuel elements to indicate by comparison with the standard under wet and dry conditions, a variation in fissile content in the absense of, or the presence of, nuclear poisons. In the prototype described in this report, provision has also been made for inserting annuli of selected materials between the fuel assembly and the reactor core to vary the moderator-reflector characteristics of the reactor and thus change the sensitivity range of the device.

Three criteria were established to guide the design of the Reactor Fuels Measurement Facility. The first basis was stated by the AEC that the installation is to have maximum mobility. Maximum mobility is to be understood as being able to move the equipment from one location to another without creating any unusual problems for a selected location. The second basis was established by the designers that the first reactor should be designed for easy modification. The reactor presented herein can be completely disassembled and all the component parts can be drastically modified. The third basis allowed personnel access to the reactor area for all conditions of reactor operation. As a result only new fuel assemblies can be handled in the installation. Adequate shielding would be required to permit handling of irradiated fuel assemblies.

5. Experimental Confirmation of the Feasibility of the Variable Loading Test Reactor Concept (12/31/59) WCAP-6040

J. A. Vreeland

A group of experiments are presented which are directed at evaluating the performance of a Variable Loading Test Reactor when used as a Reactor Fuels Measuring Facility. A parallel analytical study is presented to evaluate the effectiveness of available analytical techniques in producing comparable data.

The particular experiments performed were measurements of the sensitivity to isotopic changes in a test region moderated with clean water, borated water, and air (or unmoderated) respectively. Although the test region was quite small and the applicability of the results to large regions doubtful, the results imply the ability to identify changes as small as .01% in the fuel content of a large assembly. This ability is independent of variations in thermal or epithermal poisons which might be made in an attempt to conceal the changes in the fuel content.

The analytical results generally fall within the experimental error.

6. Assay Capabilities of a Reactor Fuels Measuring Facility (12/31/59)

WCAP-6041

J. A. Vreeland

A test reactor has been studied which is capable of assaying the content of fissionable material in a reactor fuel assembly. This assay is accomplished by means of two reactivity measurements with different loadings of fuel and moderator which make possible the separation of the effects of neutron absorption in fuel and in poison at the same neutron energies. Thus, it is possible to specify the source of any irregularity in the reactivity and to circumvent even deliberate attempts to divert fuel. Such a reactor would also be of value in studying reactor parameters in lieu of a critical experiment, and also in evaluating spent fuel for refueling or fuel cycling operations.

7. Research Contract No. AT-33-1-GEN-53 (Related to Safeguards) $ 40,000 in FY-68

Title

Decay Scheme Studies

Institution: Mound Laboratory, Miamisburg, Ohio

Principal Scientific Investigators:

Period of Contract: 1963 to (continuing)

Background & Principle

The decay scheme studies of isotopes of the actinide elements at Mound Laboratory provides the foundation to develop necessary technical skills and competence to assist programs directed toward control of fissionable materials.

The 870 Kev gamma-ray often observed from ^{238}Pu sources was at one time reported in the literature to be from the decay of ^{238}Pu (It was thought to be a gamma-ray from ^{234}U). However, this gamma-ray appeared with various intensities in gamma-ray spectra from drums containing material contaminated with ^{238}Pu. A later article in the literature suggested this was due to the ^{14}N(alpha, n)^{17}O reaction and this was confirmed at Mound Laboratory. This information was important to the ^{238}Pu assay technique developed at Mound Laboratory.

The airborne radiometric detection test of ^{238}Pu heat sources conducted at Mound Laboratory required detailed information of the neutron and gamma-ray spectra from the sources and this was supplied by Mound Laboratory. The neutron emission rates have been studied thoroughly. Earlier work at Mound Laboratory had determined that the 2.61 Mev gamma-ray observed in gamma-ray spectra from ^{238}Pu sources was due to ^{208}Tl. The results of the airborne test indicated that such sources could be located by detecting the neutrons and by detecting the 2.61 Mev gamma-ray as well as the gamma-rays known to be from the decay of ^{238}Pu.

Publications

1) Anderson, M. E., and Neff, R. A., "Neutron Emission Rates and Energy Spectra of Two ^{238}Pu Power Sources", Trans Am. Nucl. Soc., 11, 1, 91 (1968).

2) Lange, R. C., Hagee, G. R., and McCarthy, J. T., "Half Life of the 0.305 Mev Level in ^{75}As", Bull. Amer. Phys. Soc. Ser II-10-8, p. 1140 (1965), also J. Inorg. Nucl. Chem., 28, 651 (1966).

3) Hagee, G. R., Lange, R. C., and McCarthy, J. T., "Weak Alpha and Electron-Capture Decay in ^{208}Po and ^{209}Po", Nuclear Physics, 84, 62 (1966).

4) Lange, R. C., Hagee, G. R., and McCarthy, J. T., "Nuclear Levels in ^{223}Ra Populated by Alpha Decay of ^{227}Th and Beta Decay of ^{223}Fr", Bull. Amer. Phys. Soc. Ser II-11-3, p. 408 (1966).

5) Lange, R. C., Hagee, G. R., and McCarthy, J. T., "Nilsson Assignments and Rotational Structure in ^{227}Ac", Bull. Amer. Phys. Soc. Ser II-12-4, p. 521 (1967).

6) Hagee, G. R., Lange, R. C. and Campbell, A. R., "Transitions in ^{227}Ac Populated by Alpha Decay of ^{231}Pa", Bull. Amer. Phys. Soc. Ser II-12-4, p.521 (1967).

7) Hagee, G. R., Lange, R. C., Barnett, A. G., Campbell, A. R., Cothern, C. R., Griffing, D. F., and Hennecke, H. J., "The Ground State Parity of ^{227}Ac", accepted for publication in Nuclear Physics

8) Lange, R. C. and Hagee, G. R., "Levels, Transitions and Rotational Structure in ^{227}Ac", to be published in Nuclear Physics

9) Barnett, A. G., Campbell, A. R., and Hagee, G. R., "Multipolarities of Several Transitions in ^{227}Ac", to be published in Journal of Inorganic and Nuclear Chemistry.

8. Research Contract No. $

Title

Fissile Material Content by Reactivity Change-Unirradiated Fuel

Institution: Institute fur Neutronenphysik and Reactorphysik, Karlsruhe

Principal Scientific Investigators:

Period of Contract:

Objective

The possibility of measuring isotopic composition and fissile material content by reactivity change is being considered.

L. Sampling

Page

1. Liquid Samplers for Safeguards Use (USAEC) 261

2. The Mixing and Sampling of Enriched U-235 Fluids in Cylindrical Storage Containers (USAEC) 262

1. Liquid Samplers for Safeguards Use (10/1/59) WCAP-6025

W. E. Foster

Systems designed to obtain representative samples from various points in nuclear fuels reprocessing plants have been evaluated. From these sampler systems, two designs were chosen for use in a Nuclear Materials Control System (NMCS). For radioactive samples coming from feed tanks and waste hold-up tanks, the Thorex sampler was chosen. For samples coming from the product end of the plant, the Savannah River vacuum sampler system was specified.

Pertinent features concerning these sampler systems are described in the report, together with specifications for tamper-resistant features. Samples from the feed tanks and the waste hold tanks are inherently tamper-resistant because of their high radioactivity.

The specified accountability points are considered to be sufficient for reasonable safeguarding of nuclear materials in a chemical processing plant. In a disarmament situation, more accountability points would be added to those indicated in this report.

2. Research Contract No. W-7405-eng-26 $ 110,000

Title

The Mixing and Sampling of Enriched U-235 Fluids in Cylindrical Storage Containers (1/17/67)

Institution: Union Carbide Corporation

Principal Scientific Investigator: J. E. Harrell

Period of Contract: July 1965 to January 1967

Background & Principle

Solutions containing fissionable material which are generated in a fuel processing plant are stored in safe geometry cylindrical tankage and can be representatively sampled for fuel inventory after thorough mixing of tank contents.

A research program requested by the Division of Nuclear Materials Management of the United States Atomic Energy Commission for a study of the mixing and sampling characteristics of cylindrical nuclear safe-geometry tankage has been undertaken at the Y-12 Plant. A program combining the measurement of some safe-tank mixing characteristics with a theoretical analysis for a generalization of these characteristics is needed.

Procedure

For simulation of both vertically and horizontally orientated tankage, an experimental facility equipped with analytical components for flow measurement and solution-concentration measurement was assembled. Data were collected for recirculation-mixing experiments conducted in twelve tank systems of different geometric similarity and configuration. These data, a defined mixing time and the dispersion intensity, have been treated by least-squares techniques and are presented graphically as a function of the recirculation rate. For six-tank systems, the effect of the degree of fullness upon these data was observed, treated, and presented in the same manner. Both mixing-time and dispersion-intensity data were found to be linearized when plotted as a function of the recirculation rate on full logarithmic paper.

Results

The intermixing of fluid contained in cylindrical safe-geometry tankage, as found in nuclear fuel processing plants handling solutions of fissionable material, has been investigated. Utilizing an approximation to pulse-type tracer injection methods, experimental data were collected for four and six-inch-diameter single tank loops, two-in-parallel tank loops, and complex-flow-path tank loops mixed by pump recirculation, and for a six-inch-diameter single vertical tank mixed by air sparging.

A defined mixing time (mixing period needed to dispose a tracer pulse to near homogeneity with tank-loop contents) was measured for each type system and found to vary linearly with the recirculation rate when plotted on full logarithmic paper. For single tank loops, mixing time for both completely and partially fill conditions ranged between 10 and 625 minutes for recirculation rates between 1 and 100 gpm. The nature of mixing in parallel-tankage loops was considered by varying the ratio of flow through two identical tanks at aggregate flows between 10 and 100 gpm; an optimum flow split ratio where mixing time is minimized was found to exist at about 0.7. Some of the mixing characteristics for series-parallel flow through a single tank loop were studied; mixing time data for several loop systems were analyzed. In addition, the relationship between dispersion intensity which embodies the effective rate of mixing and Reynolds number was studied for several single tank loops. The relationship was found to be linear when plotted on full logarithmic paper and to deviate significantly at low Reynolds numbers from published data for very long pipelines.

Preliminary mixing-time data obtained for the air-sparged tank varied between 11 and 31 minutes at air rates between 0.12 and 5.0 scfm.

A theory for the recirculation mixing of fluids contained in cylindrical-tankage loops was developed based on the dispersion model and on the tanks-in-series model. Because either an analog or a digital computer solution is required for each theoretical treatment, only the single-tank-loop theory has yet been tested with experimental data. For this type of loop, when using an experimentally measured dispersion intensity and at Reynolds numbers greater than about 10,000, the predicted mixing times were found to agree within ± 30%.

A theoretical analysis of safe tank mixing has been made which provides a tool for treating both simple and complex tankage systems. By a combination of the series of ideally mixed tanks model and the dispersion model, mixing time for single horizontal tanks has been predicted; a comparison of the predicted times with some of those determined by experiment has also been included. Equations for the prediction of mixing time for multiple-parallel tankage and for combinations of parallel and series tankage have been developed and are outlined.

V. SPONSORING ORGANIZATION INDEX

	contracts or projects
A. ACDA	5
B. BMwF	12
C. IAEA	23
D. USAEC	82
	122

* * *

A. ACDA - Arms Control and Disarmament Agency	Page
1. Safeguards Applications of Precision Mass Spectrometry	105
2. Development of Safeguards Instruments and Techniques	115
3. Minor Isotopes Safeguards Techniques	191
4. Tamper-Resistant Instrumentation System	207
5. Tamper-Resistant Cables for Remote Monitoring	209

B. BMwF - Bundesministerium für wissenschaftliche Forschung

Page

1. Plutonium Content by Calorimetry 18
2. Isotopic Composition Analysis by X-ray Fluorescence 31
3. Isotopic Composition Analysis by Isotopic Dilution Spectrometry 32
4. Isotopic Composition Analysis by Photometry 33
5. Isotopic Composition by Delayed Neutrons-Unirradiated Fuel 101
6. Plutonium and Uranium Content in Fresh Fuel Elements Using Slowing-Down Time Techniques 107
7. Plutonium and Uranium Content in Unirradiated Fuel Pins 108
8. Mathematical Model of Fuel Cycle 176
9. Projekt Spaltstofffusskontrolle 178
10. Fissile Material Flow Control at Strategic Points in a Reprocessing Plant 179
11. Use of Statistical Analysis for the Establishment of Material Balance in a Reprocessing Plant 180
12. Fissile Material Content by Reactivity Change-Unirradiated Fuel 257

C. IAEA - International Atomic Energy Agency

			Page
1.	1958 France 4/RB $25,000	The Development of a Method of Nondestructive Analysis to Determine the U-235 and Pu Content of Irradiated Fuel Elements That Are in Storage Under Water and That Have Been Out of the Reactor for Periods of More Than One Month	53
2.	1958 Belgium 5/RB $8,000	Nondestructive Method for Evaluating the U-235 and Pu Content of Irradiated Fuel Elements	54
3.	1958 U.S.A. 7/RB $3,500	Research on the Nondestructive Analysis of Irradiated and Unirradiated Fuel Elements for Nuclear Reactors	56
4.	1959 Poland 41/RB $5,200	Nondestructive Analysis of Irradiated Fuel Elements Using a Flux Integrating Monitor	57
5.	1959 Norway 47/RB $16,000	Nondestructive Analysis of Irradiated Fuel Elements by Gamma Ray Scanning	59
6.	1960 Italy 87/RB $17,000	Development of a Method of Nondestructive Analysis of Irradiated Fuel Elements for Uranium-235 and Plutonium Content by Monitoring and Spectrometry	61
	1961 Italy 87/R_1/RB $5,000	(Extension of 87/RB from 1960)	
7.	1960 India 92/RB $30,000	A Feasibility Study of Nondestructive Assay of Plutonium-239 in Irradiated Fuel Rods Using Slowing Down Time Spectrometer	64

8.	1961 Austria 121/RB $55,000	Destructive Analysis of Unirradiated and Irradiated Fuels	109
9.	1961 Yugoslavia 129/RB $9,320	Determination of the Tritium Build-up in a Heavy Water Moderated Reactor	37
10.	1962 Sweden 163/RB $19,000	Nondestructive Analysis of Irradiated Fuel Elements by Establishing a Reliable Correlation Between One or More Gamma Lines From the Fission Products and the Burn-up of Irradiated Fuel Elements or Samples	66
	1963 Sweden 163/R_1/RB $10,000	(Extension of 163/RB from 1962)	
11.	1962 Austria 180/RB $32,700	Nondestructive Testing of Fuel Elements by Means of Compton Recoil Spectrometry	69
	1963 Austria 180/R_1/RB $16,100	(Extension of 180/RB from 1962)	
12.	1963 U.S.A. 244/RB $50,000	Development of a Photographic Identification System for Fuel Elements	193
13.	1964 U.S.A. 298/RB $30,000	Methods for Estimating the Integrated Thermal Power Produced in a Nuclear Power Reactor	225
14.	1964 U.K. 299/RB $14,020	Feasibility and Design Study of a Method to Record the Quantity of Fuel Charged and Discharged From Nuclear Reactors	228
15.	1964 Austria 314/RB $7,700	Service Contract--Analysis of Irradiated Fuel Plate from BGRR	110

16.	1966 Canada 467/CF Cost-Free	Theoretical Analysis of Plutonium Buildup and Uranium Depletion in Power Reactors	43
17.	1966 U.K. 468/RB $7,500	Theoretical Analysis of Plutonium Buildup and Uranium Depletion in Power Reactors	45
18.	1967 USSR 495/RB $30,000	Theoretical Analysis of Plutonium Build-up and Uranium Depletion in Pressurized Water and Boiling Water Power Reactors	48
19.	1966 Czechoslovakia 496/RB $19,000	Development of Safeguards Procedures for Heavy Water Moderated Gas-Cooled Reactor of Pressure Vessel Type With Continuous Refueling	133
20.	1967 U.S.A. 513/RB $31,000	Theoretical Analysis of Plutonium Buildup and Uranium Depletion in Pressurized Water and Boiling Water Power Reactors	49
21.	1967 Canada 519/RB $23,000	Development of Safeguards Procedures for Heavy-Water Moderated and Cooled Power Reactors with Continuous Refueling	135
22.	1967 South Africa 567/RO $5,000	Integrated Power Monitor for Safeguards	231
23.	1967 Canada 577/RO $20,000	Development of Safeguards Technical Practices for Pressurized Water Power Reactors	137
	$489,040		

Note: Contracts awarded by the IAEA are identified by a number and two letters, such as 468/RB or 567/RO. The numbers are assigned chronologically. The letters are for budget identification purposes: RB means Regular Budget, CF means Cost Free. For recent contracts the "O" of RO indicates the first year of the contract, while .../R1 would indicate the second year of the contract, both being supported from the regular budget.

D. USAEC - U. S. Atomic Energy Commission

Page

A. Analytical Methods

1. Selected Measurement Methods 9
2. Interlaboratory Evaluation Program 10
3. Standard Reference Materials 11
4. Analytical Procedures and Facilities for a Nuclear Materials Control System 12
5. Gamma Scan Measurement of Air Filters 13
6. Development of Primary Standards for Plutonium + Uranium 14
7. Calorimetric Assay of Radioactive Materials 15
8. Plutonium Nitrate Shipping Solution Measurements 19
9. Plutonium Oxide Shipping Measurements 21
10. Plutonium Compound Evaluation Program 24
11. Analyst Performance on Synthetic Purex Solutions 25
12. Qualification of Umpire Laboratories 26
13. Rapid Process Control of Alpha Emitting Isotopes 34

B. Calculation of Heavy Isotope Content of Reactor Fuel

14. Analytical and Experimental Methods of Determining Heavy Isotope Content of Operating Fuel Elements 39

C. Fuel Element Analysis

15. A Gamma-Ray Scanner for Safeguards Use 71

16. The Fuel Assay Scanner 72

17. Field Testing of the Fuel Assay Scanner 73

18. A Method for the Nondestructive Analysis of Power Reactor Fuel Elements 74

19. Nondestructive Assay of Nuclear Fuel 75

20. Nondestructive Analysis of Burnup in Irradiated Fuel Elements 77

21. Pulsed Neutron-Research for Nuclear Safeguards 78

22. Application of Photoinduced Reactions to Nuclear Safeguards 90

23. Laser Microprobe Analyser 96

24. High-Resolution Gamma Spectrometry for Analysis of Irradiated Fuel 98

25. Gamma-Ray Assay of Pu-238 in Waste Drums 102

26. In-Line Gamma-Ray Assay of Pu-238 in Waste Cans 103

D. Gamma Spectrometers

27. Portable Gamma Spectrometer 113

28. Portable Single Channel Gamma Spectrometer 114

E. Inspection Procedures

29. Nuclear Materials Control System 119

30. Formulation and Evaluation of Inspection Procedures for the Application of Safeguards to the Yankee Nuclear Power Station 124

31. Application of Safeguards to Nuclear Fuel Processing Plants 125

32. Application of Safeguards to Fuel Fabricating Plants 128

33. Safeguards Procedure Manual for the NFS Reprocessing Plant 131

34. Evaluation of Resident Inspection Trial Program 139

F. Inventory Control

Page

35. Inventory Control Study 145

36. Statistical Methods to Evaluate Material Control 146

37. Nuclear Materials Control Manual 150

38. Evaluation of Methods of Input Accountability in a Reprocessing Plant 152

39. Standardization of AEC Inventory Verification Procedures 157

40. Material Accountability in Fluidized Bed Processes 161

G. Mathematical Models for Fuel Cycle Analysis

41. Mathematical Models and Decision Functions Study 165

42. Systems Studies for Special Nuclear Material Safeguards 167

43. Nuclear Process Analysis Studies 171

H. Miscellaneous

44. Technical Support Organization 185

45. Investigation of the Radiation Chemistry of Plutonium Nitrate Solutions 195

46. Fuel Sample Stability 199

47. Evaluation of Computers for Nuclear Materials Control 202

48. Radioactive Waste Containers for Storage and Shipping 203

49. Bilateral Certification of Reactor Design 204

I. Plant Protective and Monitoring Systems and Devices

50. Tamper-Indicating Safing System 210

51. Personnel Monitoring to Detect Diversion of Fissionable Materials in Chemical Processing Plants 211

52. Closed Circuit Television for Security and Surveillance 212

53. Devices and Techniques for Perimeter Protection 213

54. Devices and Techniques for Protection of Enclosed Spaces 214

55. Devices and Techniques for Access Control and Protection 215

56. Devices and Techniques for Protection of Pipe and Conduit 216

57. Alarm Transmitting Systems and the Protection of Communications 217

58. Miscellaneous Devices for Plant Security 218

59. Miscellaneous Devices for Safeguarding a Fuels Reprocessing Plant 219

60. General Activity Detectors for Safeguards Use 220

61. A Doorway Monitor for Safeguards Use 221

J. Process Instrumentation

62. Metrology of Nuclear Materials 232

63. In-Line Monitor for Fissionable Material 235

64. Volume and Density Measurements Using Ashcroft Pressure Gages 237

65. Volume and Density Measurements on Radioactive Solutions 238

66. Recording Wattmeters for Safeguards Use 239

67. Neutron Flux Monitors for Safeguards Use 240

68. A Thermal Power Meter for Safeguards Use 241

69. Continuous Monitors for Uranium and Plutonium in a Chemical Processing Plant 242

70. Specific Gravity Instruments for a Nuclear Fuels Reprocessing Plant 243

71. Liquid Level Instruments for a Nuclear Fuels Reprocessing Plant 244

72. Flowmeters for a Nuclear Fuels Reprocessing Plant 245

73. Evaluation of N-16 Monitoring in WTR Primary Coolant for Measurement Flow and Reactor Power 246

K. Reactivity Measurements

74. Preliminary Considerations in the Design of the Westinghouse Variable Loading Test Reactor 249

75. The Feasibility of an Annular Core Reactor as a Reactor Fuels Measuring Device 250

76. The Feasibility of an Annular Nuclear Core as a Reactor Fuels Measuring Facility 251

77. A Design for the Evaluation of a Reactor Fuels Measurement Facility 252

78. Experimental Confirmation of the Feasibility of the Variable Loading Test Reactor Concept 253

79. Assay Capabilities of a Reactor Fuels Measuring Facility 254

80. Decay Scheme Studies 255

L. Sampling

81. The Mixing and Sampling of Enriched U-235 Fluids in Cylindrical Storage Containers 262

82. Liquid Samplers for Safeguards Use 261

VI. USAEC CONTRACTS FOR RESEARCH RELATED TO SAFEGUARDS

In addition to research contracts awarded specifically for either nuclear materials control or safeguards, the USAEC has supported other work which is related to safeguards. Typical examples of such work are projects conducted at the Mound Laboratory which is a prime contractor to USAEC.

It must be recognized that many research programs are "related" to safeguards in that the results contribute to the broad areas of technology upon which safeguards depends. The best source of information for such related work which is not specifically funded for safeguards is "Nuclear Science Abstracts."

The USAEC supported work at Mound Laboratory which may be considered closely related to safeguards is as follows:

		Page
1.	Development of Primary Standards for Plutonium + Uranium	14
2.	Calorimetric Assay of Radioactive Materials	15
3.	Rapid Process Control of Alpha Emitting Isotopes	34
4.	Gamma-Ray Assay of Pu-238 in Waste Drums	102
5.	In-Line Gamma-Ray Assay of Pu-238 in Waste Cans	103
6.	Radioactive Waste Containers for Storage and Shipping	203
7.	Volume and Density Measurements Using Ashcroft Pressure Gages	237
8.	Volume and Density Measurements on Radioactive Solutions	238
9.	Decay Scheme Studies	255

VII. EXPENDITURES FOR SAFEGUARDS RESEARCH

Both the USAEC and the IAEA award contracts at any time of the year. In order to get a more realistic picture of the actual expenditures by year, the total price for each contract was divided into expenditures by six month periods. Thus the calendar year totals below represent the sums of the six month expenditures for all contracts being reported.

	USAEC	IAEA	ACDA
1955	61,500	--	--
1956	71,500	--	--
1957	288,574	--	--
1958	647,446	--	--
1959	881,321	20,000	--
1960	707,446	27,967	--
1961	328,571	23,066	--
1962	340,625	45,493	--
1963	124,221	81,494	--
1964	235,719	90,042	--
1965	343,054	59,478	--
1966	478,382	7,875	--
1967	945,832	83,125	106,000
1968*	2,087,168	43,832	275,000
	$7,541,359	$482,372	$381,000

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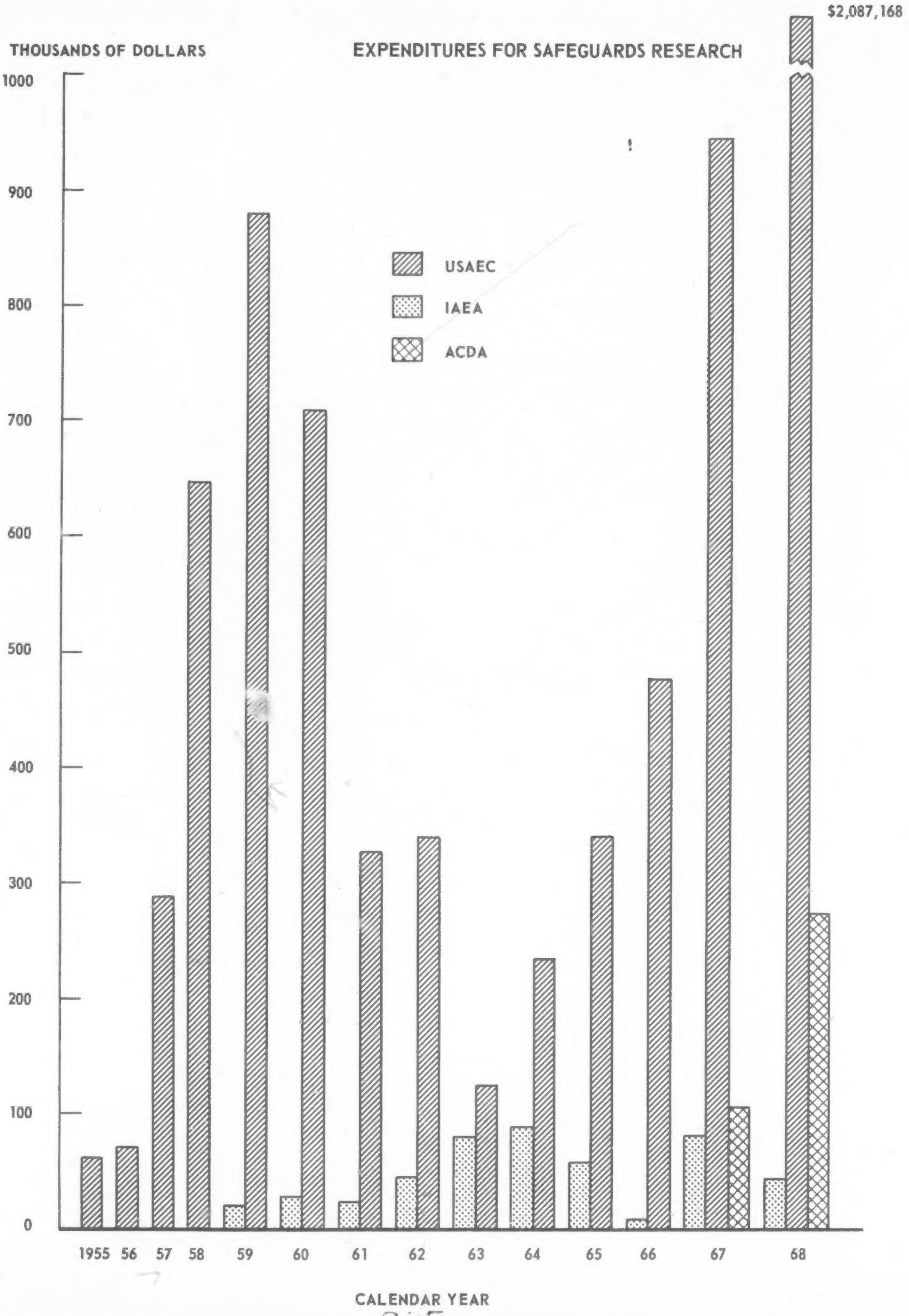
*The figures for 1968 include expenditures extrapolated to December 31, 1968.


THOUSANDS OF DOLLARS
EXPENDITURES FOR SAFEGUARDS RESEARCH
$2,087,168
1000
900
800
700
600
500
400
300
200
100
0
USAEC
IAEA
ACDA
1955
56
57
58
59
60
61
62
63
64
65
66
67
68
CALENDAR YEAR

VIII. BASIS AND BACKGROUND OF SAFEGUARDS

Definition of Safeguards

Safeguards is a collective term that comprises those measures designed to prevent and detect unlawful diversion of materials such as special nuclear material (Pu, uranium enriched in U-233 or U-235, or any material artificially enriched by any of the foregoing).

There are three general elements of safeguards:

Physical security - this includes measures such as vaults, locks, seals, guards, fences, and alarms intended to prevent diversion by providing a high probability of immediate detection. Measures to insure reliability of employees may be included in this category.

Accountability - includes measurement, accounting, auditing and inventory procedures designed to provide an accurate knowledge of material quantities and location. The system, by accounting for the nuclear material in the various segments of the nuclear fuel cycle, provides a mechanism for detection if amounts of material in excess of measurement uncertainties are diverted from the cycle.

Surveillance - or monitoring is the use by the safeguards agency of its own agents and/or monitoring instruments to obtain independent information on a periodic or continuous basis about material flow or inventory, or performance of a plant, process, security system or accountability system.

A. United States Arms Control and Disarmament Agency

"CREATION AND AUTHORITY.---The United States Arms Control and Disarmament Agency was established by the act approved September 26, 1961 (75 Stat. 631;22 U.S.C. 2551).

"PURPOSE.---The Agency is responsible for the conduct, support, and coordination of research for arms control and disarmament policy formulation; the preparation for and management of United States participation in international negotiations in the arms control and disarmament field; the dissemination and coordination of public information concerning arms control and disarmament; and the preparation for, operation of, or, as appropriate, direction of United States participation in such international control systems as may under treaty arrangements become part of United States arms control and disarmament activities.

"ACTIVITIES.---A major share of the Agency's efforts has gone into discussions and negotiations, both at the United Nations and at Geneva, on the nonproliferation of nuclear weapons, banning nuclear weapons tests, and other measures to contain the nuclear threat, as well as more general disarmament proposals. The Agency is actively engaged in efforts to control the international traffic in conventional arms. Research occupies an important role, since the Agency is responsible for insuring the conduct of research into the manifold problems of arms control and disarmament through (1) studies performed with its own resources; (2) arrangements, including contracts, agreements, and grants, for the conduct of research, development, and other studies by private or public institutions or persons; and (3) coordination of activities conducted in this field by or for other Government agencies in accordance with procedures established by the organic law."

B. BMwF - Bundesministerium für wissenschaftliche Forschung
(Federal Ministry for Scientific Research)
Federal Republic of Germany

The Federal Republic of Germany (FRG) has no large national agency for nuclear activities, since these are entirely non-military in nature. The Federal government can give financial assistance to the States, which themselves have considerable autonomy under the German system of government. Further, the economic policy has been for the Government not to compete in any way with publicly or privately owned utilities. For these reasons, the Federal Ministry for Scientific Research is limited to outlining general policies for research and development and for the use of nuclear energy. It also prepares legislation. The German Atomic Energy Commission, DAK, is essentially a large advisory committee to the ministry. Actual administration of atomic energy projects rests with the States or private industry.

The Bundesministerium für wissenschaftliche Forschung (BMwF), Federal Ministry for Scientific Research, was created in 1962 to assume the functions of the Federal Ministry of Nuclear Energy and Water Economy which, formerly, was called the Federal Ministry for Atomic Affairs. The BMwF is concerned with the promotion of all fields of scientific endeavour in Germany, nuclear energy, aeronautical, space and computer research. The BMwF has an Atomic Energy Advisory Committee (Deutsche Atomkommission: DAK) and a Bundestag Nuclear Energy Committee. The latter comprises members of parliament who are familiar with atomic and other scientific affairs. The detailed organization of German nuclear research is administered by university research institutes, the Max Planck Institutes, industry, the large scientific research institutes and the State governments.

One of the applied research centers in Germany is the Kernforschungszentrum Karlsruhe, Nuclear Research Centre Karlsruhe, which was founded in 1956. It is operated by the Gesellschaft für Kernforschung mbH, and expenses are borne 75% by the Federal Government and 25% by the Land of Baden-Württemberg.

The Centre has major facilities for research in radiochemistry and other topics of interest include: physics, radiobiology, food preservation, hot chemistry, control engineering, reactor engineering, nuclear engineering, technology, metallurgy and development of components.

The above information is from "Atomic Handbook, Vol. I, 1965, Europe."

C. International Atomic Energy Agency

On December 8, 1953, the President of the United States made a proposal before the United Nations for the establishment of an International Atomic Energy Agency (IAEA) to bring the benefits of peaceful uses of atomic energy to all the peoples of the world. The IAEA Statute, which is its governing law, entered into force on July 29, 1957. It states the objectives of the organization to be the following:

> "The Agency shall seek to accelerate and enlarge the contribution of atomic energy to peace, health and prosperity throughout the world. It shall ensure, so far as it is able, that assistance provided by it or at its request or under its supervision or control is not used in such a way as to further any military purpose." (Article II)

Among its functions the Agency is authorized:

> "To establish and administer safeguards designed to ensure that special fissionable and other materials, services, equipment, facilities, and information made available by the Agency or at its request or under its supervision or control are not used in such a way as to further any military purpose; and to apply safeguards, at the request of the parties, to any bilateral or multilateral arrangement, or at the request of a State, to any of that State's activities in the field of atomic energy." (Article III) (emphasis added).

In order to properly discharge its responsibility for the administration of peaceful use safeguards, the Agency is directed to establish a staff of inspectors. The staff of inspectors has the responsibility of verifying that facilities and materials subject to safeguards are not used in furtherance of any military purpose.

There are now 98 members of IAEA. During the 12th General Conference of the IAEA, concluded on September 30, 1968, three more countries were approved for membership.

D. United States Atomic Energy Commission

Atomic Energy Act of 1946

From the enactment of the Atomic Energy Act of 1946 until it was extensively revised in 1954, all special nuclear material remained in the hands of the United States Government or its contractors. With the exception of very small quantities of material used by some universities conducting research under contract with the USAEC, all special nuclear material was held by the USAEC and its cost-type contractors operating government-owned or controlled plants and laboratories.

Atomic Energy Act of 1954

The Atomic Energy Act of 1946 was extensively revised in 1954. Under the 1954 Act, special nuclear material was made available to private persons for peaceful uses, although private ownership of special nuclear material was not permitted until 1964. The authority for the USAEC to safeguard special nuclear material is provided by the following provisions of the Atomic Energy Act, among others:

> "Sec. 161. General Provisions—In the performance of its functions the Commission is authorized to —
>
> "b. establish by rule, regulation, or order, such standards and instructions to govern the possession and use of special nuclear material, source material, and byproduct material as the Commission may deem necessary or desirable to promote the common defense and security or to protect health or to minimize danger to life or property;
>
> "i. prescribe such regulations or orders as it may deem necessary...(2) to guard against the loss or diversion of any special nuclear material acquired by any person pursuant to section 53 or produced by any person in connection with any activity authorized pursuant to this Act, and to prevent any use or disposition thereof which the Commission may determine to be inimical to the common defense and security..."

The 1954 Act also permitted the USAEC to supply special nuclear material to foreign governments and to international organizations, such as the International Atomic Energy Agency, for peaceful purposes. The section which permits such cooperation includes a provision for safeguards against diversion of the material to non-peaceful uses:

> "Sec. 123. COOPERATION WITH OTHER NATIONS.—No cooperation with any nation or regional defense organization pursuant to sections 53, 54, 57, 64, 82, 91, 103, 104, or 144 shall be undertaken until—
>
> "a. the Commission or, in the case of those agreements for cooperation arranged pursuant to subsection 91 c. or 144 b. which are to be implemented by the Department of Defense, the Department of Defense has submitted to the President the proposed agreement for cooperation, together with its recommendations thereon, which proposed agreement shall include (1) the terms, conditions, duration, nature, and scope of the cooperation; (2) a guaranty by the cooperating party that security safeguards and standards as set forth in the agreement for cooperation will be maintained; (3) except in the case of those agreements for cooperation arranged pursuant to subsection 91 c. a guaranty by the cooperating party that any material to be transferred pursuant to such agreement will not be used for atomic weapons, or for research on or development of atomic weapons or for any other military purpose; and (4) a guaranty by the cooperating party that any material or any Restricted Data to be transferred pursuant to the agreement for cooperation will not be transferred to unauthorized persons or beyond the jurisdiction of the cooperating party, except as specified in the agreement for cooperation;" (emphasis added)

Control of Special Nuclear Material by the USAEC

Following the passage of the Atomic Energy Act of 1954, the Atomic Energy Commission established the Division of Nuclear Materials Management which, among others, had the following functions:

> "c. plans and administers programs for the development, certification and distribution of chemical and isotopic standards and for the development and selection of standard measurement methods."
>
> "d. provides staff guidance and assistance in the implementation of the control procedures in effect for source and special nuclear materials."
>
> "e. provides the Divisions of Licensing and Regulation and International Affairs with staff guidance on reporting and control systems for source and special nuclear materials distributed under programs of their respective divisions." (AEC 0103-26 Approved: 10/21/60)

Under the above authority, the Division funded contracts for the development of accounting procedures, fuel burnup calculations and analytical methods.

On April 5, 1967, the Commission approved the establishment of the Office of Safeguards and Materials Management and simultaneously abolished the Division of Nuclear Materials Management, These organizational changes were effective July 1, 1967. Among the functions assigned to the new office is the following:

> "e. identifies need for and conducts research and development in support of AEC safeguards and materials management programs; establishes, and provides material for, measurement standards for SS materials." (AEC 0124-01 Approved: 2/23/68)

☆ U.S. GOVERNMENT PRINTING OFFICE : 1968 O—327-227